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A study of the Roman iron bars of Saintes-Maries-de-la-Mer (Bouches-du-Rhône, France). A proposal for a comprehensive metallographic approach

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ABSTRACT

The aim of this paper is to contribute to the understanding of the organisation of the manufacture and circulation of iron bars and the possible bar standardisation of a given set of artefacts in order to throw more light on commercial patterns during Antiquity. A set of 48 iron based metal bars originating from the Roman shipwrecks at Les Saintes-Maries-de-la-Mer (France) has been studied using comprehensive metallographic observations, ranging from macroscopic to microscopic scales and slag inclusion (SI) analyses. A comparison of the results allows one to distinguish different metal qualities (inclusions, pores, welding, carbon content) that may be linked to morphological types, which shows the possible standardisation of this set of artefacts. Moreover, SI analyses allow distinguishing various origins for the Primary Pieces of Metal, thereby throwing light on a specific organisation of the iron bar production line during the late Roman period.

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1. Introduction

The understanding of commercial practice, linked to a specific historical and technical context, is one of the crucial issues of history and archaeology. Iron based materials have been significant components of the commercial landscape since Antiquity. A better overview of the form and modality of circulation and diffusion of such materials, and especially of the half-products that are found at several archaeological sites, is therefore a key issue.

Because of the variable nature of the ancient metal structure, it is necessary to employ a specific methodology for the study of archaeological ferrous alloys. Since the post-war years, metallography has been regularly used in archaeological studies. A thorough discussion has contributed to the development of an appropriate archaeometallurgical methodology that serves to explain the observed features of different artefacts (Salin, 1952–1959; Forbes, 1955, 1963; 1964; France-Lanord, 1963; Scott, 1971; Piaskowski, 1984a,b; Pleiner, 1984; Fluzin et al., 2000, 2004). However, the majority of the metallographic examinations of small or large objects have only been performed at the microscopic level. In this paper, we propose to follow this train of thought, and to do so especially by taking into account both macroscopic and microscopic observations, as advised by Pleiner (1984).

Hence, considering all these aspects, the main aim of the present paper is to try to inform the organisation of the manufacture of iron bars and the condition of circulation of a given set of artefacts in order to provide more information on the commercial patterns during Antiquity. This will be carried out by performing archaeological, metallographic and chemical analyses on the well-known collection of bars originating from Roman shipwrecks that have been discovered off the coast of Saintes-Maries-de-la-Mer (south of France), in the Bouches-du-Rhône, by Luc Long (Long et al., 2002, 2005; Long, 2003; Long and Sintes, 2003) in the framework of a project conducted by the DRASSM (Département des Recherches Archéologiques Subaquatiques et Sous-Marines, Marseille, Bouches-du-Rhône, France).

The artefacts analysed here originate from eleven Roman ships containing only ferrous bars (from 20 to 150 tons, according to the size of the ships and the vestiges found). The total weight of the eleven wrecks is approximately 500 tons. This estimated mass is intentionally underestimated, but gives an idea of the order of magnitude in question. Indeed, it is very possible that part of the freight was recollected after the shipwreck because, at the time, the wreck area was located only several meters from the shore. The

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eleven Roman ships were used during the 1st century AD (more precisely between 27 BC and 96 AD) in a former Rhône branch: the Saint-Ferréol Rhône (Fig. 1). Archaeological evidence leads to the hypothesis that these ships were meant to go up the Saint-Ferréol Rhône from the Mediterranean, or at least to cast anchor in front of this former Rhône branch in order to transfer their cargo on to other ships, adapted to fluvial conditions (Long et al., 2002, 2005; Long, 2003; Long and Sintes, 2003; Coustures et al., 2006; Pagès et al., 2008; Pagès, 2010). This shipwreck collection is exceptional in terms of its state of conservation, richness, contemporaneousness and readability. It gives a snapshot both of the marine circulation and of the ferrous bars that were used during the period of the rise of the Roman Empire in the Mediterranean.

First, a selection of bars, representative of each different types, have been analysed following a systematic metallographic methodology in order to identify possible trends and links between the microstructure of the metal and the morphology of the bars. Then, using major element analysis of slag inclusions (Dillmann and L'héritier, 2007; Blakelock et al., 2009), the variability of the reduction systems used to obtain the metal of the different products was considered and discussed. Actually, this point is crucial because in previous studies (Coustures et al., 2003), trace element analysis of slag inclusions by LA-ICP-MS led the authors to suggest that two bars (types 4C and 1L), respectively originating from the wrecks SM9 and SM10 of Saintes-Maries-de-la-Mer, were compatible with a source from the Montagne Noire (Aude, France). This hypothesis was actually proposed for a long time, taking into consideration archaeological and epigraphic data (Solier, 1981; Sablayrolles, 1989; Domergue, 1993; Decombeix et al., 1998; Long et al., 2002). Nevertheless, in the beginning of the Roman period, metal production saw a significant increase in the entire territory of Gaul (Domergue et al., 2006). In the surrounding Mediterranean area, this development was observed in the Montagne Noire, but also in the Canigou area (Oriental Pyrenees), in the Hautes Corbières and in Spain (Rancoule and Solier, 1977; Barrouillet et al., 1989; Cambon, 1989; Izard, 1994; Pauc and Pauc, 1998; Rico, 2005; Mut and Kotarba, 2007). Considering these aspects, even if part of the iron making up the bars of the Saintes-Maries-de-la-Mer wrecks originates from the Montagne Noire, it is also probable that a large part of the bars found in the wrecks comes from other iron smelting areas. As it is not possible today to determine the chemical signature of each of these potential production areas and to answer the question of origin with certainty, it appears that the bars of Saintes-Maries-de-la-Mer can be analysed so as to address another issue: how was the iron trade organised at the beginning of the Roman period? This can be assessed firstly by combining and confronting the results of metallographic observations, taking into account the complexity of the bars and major element slag inclusion analyses. Then, the technical and economic aspects entailed by the manufacturing of the bars of Saintes-Maries-de-la-Mer will be considered in the discussion part.

Table 1
Metrological characteristics of Saintes-Maries-de-la-Mer bars.

Type	Length (mm)	Width (mm)	Thickness (mm)	Weight (g)
1L	900–1400	35–50	15–25	8000–11000
1M	500–700	35–50	15–25	2000–4000
2M	400–700	20–40	20–40	2000–5000
3C	300–400	35–45	30–40	1500–4500
4C	200–300	45–75	35–65	2000–7000
4L	1000–1900	45–75	35–65	22000–33000
5C	250–350	45–55	30–40	2500–3500
6C	250–350	90–120	30–50	4000–8000

2. Artefacts and analytical methodology

2.1. Analysed artefacts

Eight morphological bar types were identified in previous studies (Coustures et al., 2006; Long et al., 2005) based on two

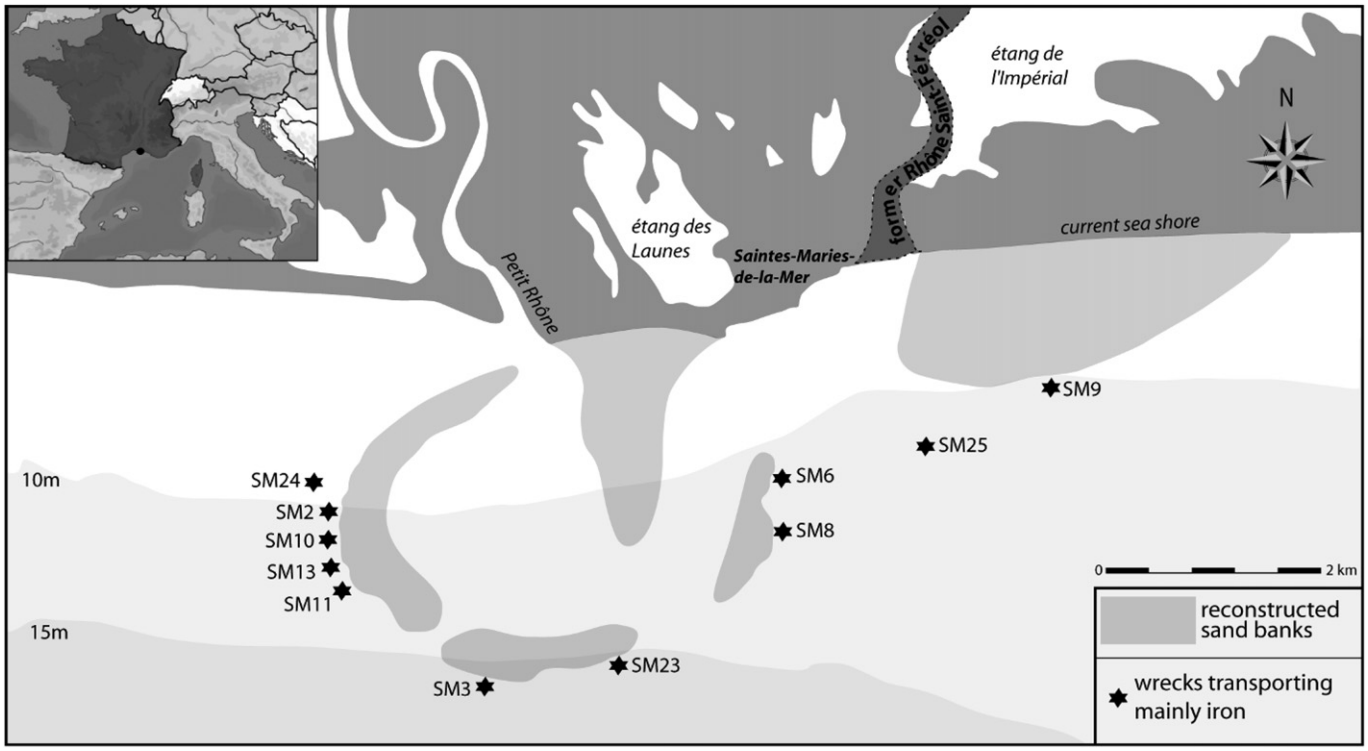

Fig. 1. Location of Saintes-Maries-de-la-Mer shipwrecks containing ferrous bars in the Bouches-du-Rhône (south of France).

metrological criteria: the section area (arbitrarily identified by a number) and the length (indicated with a letter – C: short, M: medium, L: long). These typologies are presented in Table 1 and Fig. 2. Lengths between 0.2 and 2 m and Weights range between 1.5 and 33 kg. Types 1L, 2M and 4C are the most common.

Because 3C and 5C types are very rare in the different wrecks, only the other six ones were studied here. These types of bars were collected from different wrecks, as indicated in Table 2. Forty-eight bars were taken from six of the eleven excavated wrecks. After *ad hoc* preparation (see below), all these bars were studied at a macroscopic scale, and twenty of them were studied at the microscopic scale. Since their length is considerable, microscopic scale studies were performed at different places on the same bar, on thirty-five samples (Table 2).

2.2. Metallographic observations

The heterogeneous structure of the bars requires to continuously switch between a complete macroscopic observation and microscopic scale examinations in order to reach a representative vision of the artefact. For this purpose, a specific methodology was set up.

The first step consisted of cutting or eroding the surface in order to reach the non-corroded core of the bar. This was carried out using a milling machine or high pressure water cutting. A main side and a lateral perpendicular side of the bar were ground (grade 180) all along their entire length. It was then possible to observe, at macroscopic scale, the metal in 'three dimensions'. After Nital 4% etching (between 15 and 20 s), the welding lines appeared as darker lines at this level of observation (Fig. 3). Moreover, after Nital etching, areas of the surface that presented carbon contents higher than 0.2–0.3 wt% appeared darker than the areas with lower carbon contents, which remained bright (Fig. 3). This allowed us to distinguish the more carburised zones at this macroscopic scale and evaluate by image analysis (using Adobe Photoshop) the proportion of the surface that can be considered as 'steely' (i.e. over 0.2–0.3 wt% carbon).

The second phase of the analytical protocol consisted of taking representative samples of each bar (of about 20 cm long) and polishing them down to 1 μm (using diamond paste) in order to carry out microscopic metallographic observations (using a Leica optical microscope – magnification up to 1000×). If the length of the bar was greater than 30–40 cm, it was necessary to take more than one sample (Figs. 4 and 9–11).

This stage allows a fine study of the metal cleanliness (quality and quantity of inclusions, pores, etc.) by image analysis. After the Nital etching, the microstructure of the samples was made visible, together with the welding line locations, as well as the carbon content and distribution. This latter parameter was evaluated by quantitative metallography.

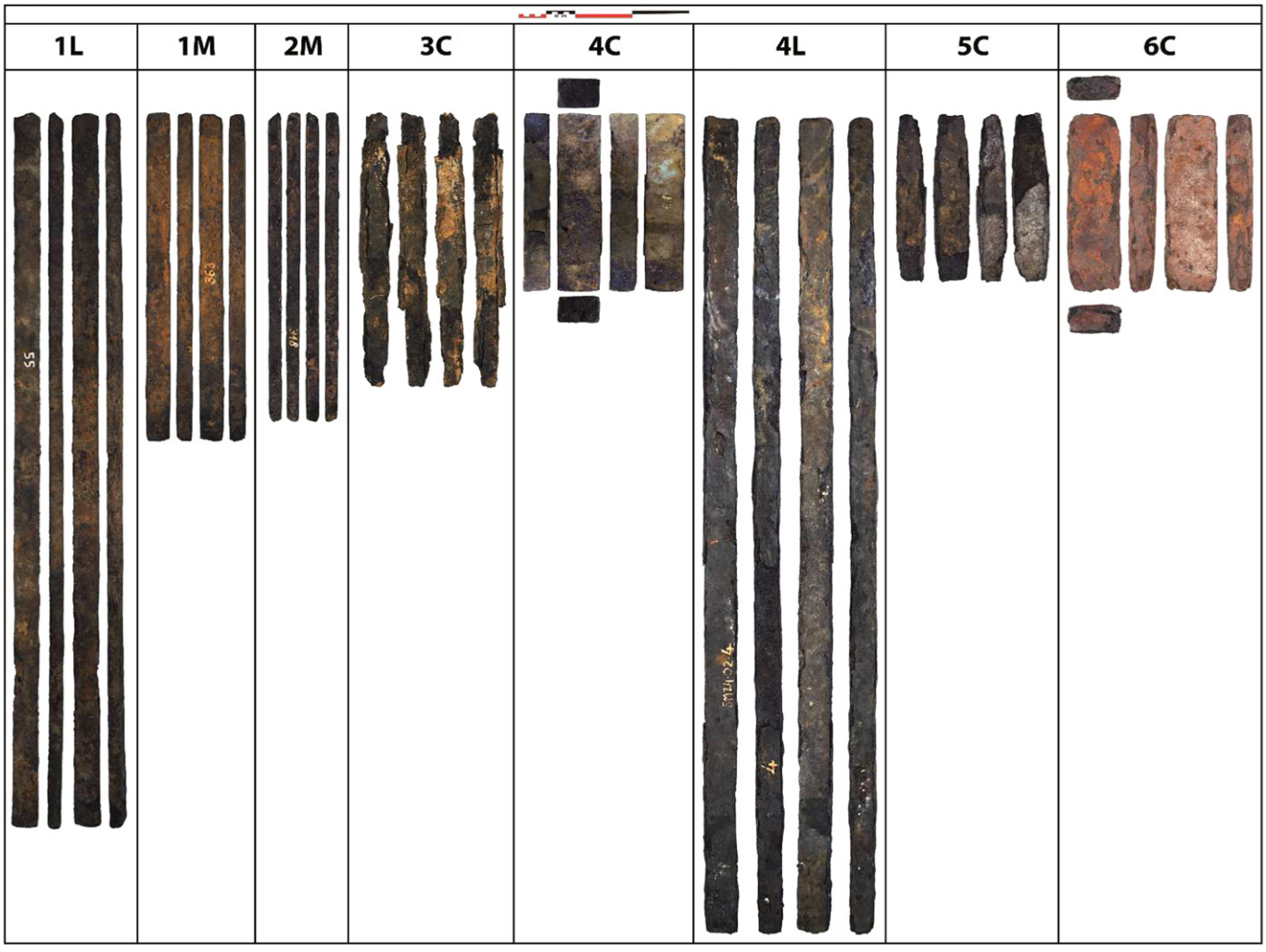

Fig. 2. General aspects of the various types of Saintes-Maries-de-la-Mer bars.

Table 2
The analysed bars from Saintes-Maries-de-la-Mer. Bar identification: type, wreck number, bar number.

Bar identification	Length (mm)	Width (mm)	Thickness (mm)	Weight (g)	Macroscopic metallography	Microscopic metallography	Number of cross sections
1L SM2_1	1087	45	22	5680	X	X	2
1L SM2_2	1025	40	23	5230		X	2
1L SM2_7	1075	39	15	4480	X		
1L SM2_8	1065	38	17	4270	X		
1L SM10_1	1300	45	20	9305	X	X	3
1L SM10_2	1266	45	25	8400	X	X	3
1L SM10_3	1306	49	25	9560	X		
1L SM10_4	1170	44	23	6110	X		
1M SM9_5	665	47	25	3664	X		
1M SM9_6	610	44	27	3580	X	X	1
1M SM9_7	540	44	26	2510	X		
1M SM9_8	667	41	20	4000	X	X	1
1M SM9_9	583	42	26	4140	X	X	1
1M SM9_10	631	40	26	4582	X	X	1
2M SM6_3	470	50	45	3104	X		
2M SM6_4	433	39	31	2038	X		
2M SM9_1	512	39	38	3824	X	X	2
2M SM9_2	470	37	38	2782		X	1
2M SM9_13	483	33	31	2310	X		
2M SM25_1	600	32	32	3884	X	X	1
2M SM25_2	558	23	23	1914	X	X	2
2M SM25_3	626	32	32	4540	X		
2M SM25_4	492	32	32	3590	X		
2M SM25_5	533	25	25	1980	X		
2M SM25_6	560	26	25	2350	X		
4C SM2_3	240	54	40	1976	X		
4C SM2_4	262	62	45	4258	X		
4C SM2_5	236	55	43	3744	X	X	1
4C SM6_2	272	48	36	2838	X	X	1
4C SM6_5	280	44	58	2560	X		
4C SM6_6	266	58	39	4240	X		
4C SM6_7	264	63	43	3820	X		
4C SM9_3	260	64	47	4602	X	X	2
4C SM9_4	250	67	50	3200		X	2
4C SM9_11	297	66	42	5855	X	X	1
4C SM9_12	238	62	38	3878	X		
4C SM24_7	280	75	60	6870	X		
4C SM24_8	300	56	53	5130	X		
4L SM24_1	1160	58	40	13,950	X	X	2
4L SM24_2	1483	56	43	22,900	X	X	5
4L SM24_3	1624	66	47	27,140	X		
4L SM24_4	1358	57	43	17,870	X		
4L SM24_5	1352	47	36	13,920	X		
4L SM24_6	1015	53	29	9480	X		
6C SM6_1	320	89	43	4910	X	X	1
6C SM6_8	354	150	47	8120	X		
6C SM6_9	280	115	40	4890	X		
6C SM6_10	295	105	52	6790	X		

The surface proportion of different carbon classes was determined as follows: <0.02, 0.02–0.2, 0.2–0.4, 0.4–0.6, 0.6–0.8, 0.8–1, etc. (Table 3). For an easier quantification of the different surface proportions in the sample, schematic drawings of the carbon content and distribution were made starting from high-resolution microphotographs (Fig. 4).

A simple average measurement of the carbon content does not express the distribution of this element accurately. Therefore, we decided to define a weighted carbon content as follows:

$$\%C^* = \sum_{i=1}^{n} p_i \times x_i$$

where %C* is the weighted carbon content, x_i is the mean C content for a given class and p_i is the surface proportion corresponding to the given class (sum of $p_i = 1$).

After re-polishing, Oberhoffer etching showed a phosphorous content and distribution between 0.1 and 0.6 wt% (Stewart et al., 2000; Vega et al., 2003) (Fig. 11). In order to detect possible higher concentrations (over 0.5%), Energy Dispersive Spectrometry (EDS) measurements coupled with Scanning Electron Microscopy (SEM) were performed on the metallic matrix.

2.3. Slag inclusion analyses

A complete inclusion study following a procedure published elsewhere (Dillmann and L'héritier, 2007) was performed on each sample. In the present study, only major elements of the slag inclusions were analysed by EDS coupled to SEM. The measurement conditions were the same as those proposed by Dillmann and L'héritier (2007). The entire surface of each analysed inclusion was measured and taken into account. Moreover, it was verified for multi-phased inclusions by comparison with the LA-ICP-MS results, so that the error caused by the approximations for quantification were lower than the variability of the slag inclusion (SI) concentration in a given artefact. This approach does not aim to determine the origin of the metal, but permits distinguishing, in some favourable cases, different reduction systems (i.e. ore, lining, charcoal and additive). It can be assumed that, in the technical context

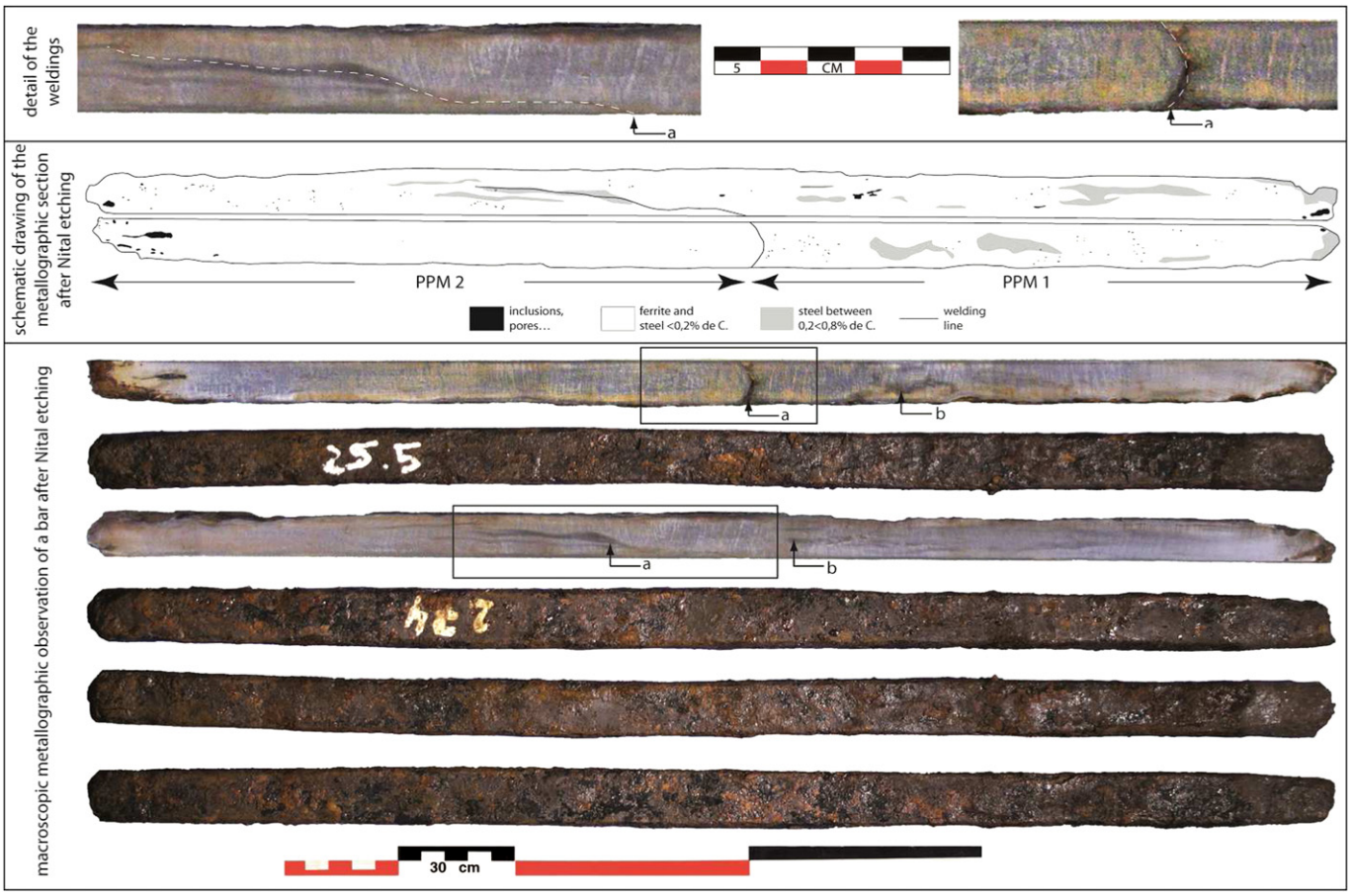

Fig. 3. An example (sample 2M SM25_5) of a macroscopic metallographic observation of a bar after Nital etching. (a) Welding lines; (b) Surface that presents carbon contents higher than 0.2–0.3 wt%.

studied here, in a given period of time corresponding to the manufacturing of a set of bars, a workshop would have employed the same practices, linked to the environment (i.e. raw materials available) and used the same combination of these elements. This study is a necessary and a complementary first step to other analyses dealing with the origins of the metal which take into account trace elements (Coustures et al., 2003, 2006; Desaulty, 2008; Desaulty et al., 2009; Leroy, 2010) or isotopes (Schwab et al., 2006).

To identify inclusions originating from intentionally added compounds during the forging stage, but also local enrichment effects mainly due to the small sizes of the inclusions, in addition to metallographic examinations (for locating welding lines for example), it is necessary to analyse a large number of inclusions per artefact (at least 40) and to then proceed to filtering the raw results. For this purpose, only Non Reduced Compounds (NRC) are considered (i.e. Al_2O_3, SiO_2, K_2O, MgO and CaO). The Al_2O_3 versus SiO_2, K_2O versus CaO and MgO versus Al_2O_3 diagrams are plotted and it is verified that most of the inclusions present constant NRC ratios (i.e. could be modelled by a line passing through zero with a good determination coefficient) and can be associated to the reduction stage. Inclusions that do not conform to this pattern (either because they do not originate from the reduction stage, are polluted by additives, or because of local concentration effects), are not considered. If, following this procedure (for more details see Dillmann and L'héritier, 2007), a determination coefficient over 0.6 can be obtained for a given sample and for each considered NRC ratio, then a weighted content for all elements (including the non-NRC) can be calculated. This weighted content is estimated by taking into account the ratio between the surface area of the analysed inclusion and the total analysed surface:

$$\%E^* = \sum_{i=1}^{n} \left(\%E_i \times \frac{S_i}{S_T} \right)$$

where $\%E^*$ is the weighted content of the considered element or oxide, $\%E_i$ is the mass content of the element or oxide in the i Slag Inclusion (SI), S_i is the surface of the SI where the analysis is performed, S_T is the total surface of the analysed SI, and n the total number of inclusions. This weighted content is indicated in the following with an * in order to distinguish it from the measured contents (for example $\%Al_2O_3{}^*$). It can be related to a pseudo-macroscopic average content and thus be compared to the macroscopic slag composition.

Moreover, to compare the contents of the elements that are not NRC, especially MnO and P_2O_5, and in order to eliminate the effect of the reduction conditions in the furnace, the weighted compositions were normalised at 100% without FeO (see columns 10 and 11 in Table 5).

3. Results

For the periods under consideration here, when iron based artefacts are voluminous, they are often made up of one or several Primary Pieces of Metal (PPM), each one originating from an ore reduction operation performed by the bloomery process. These

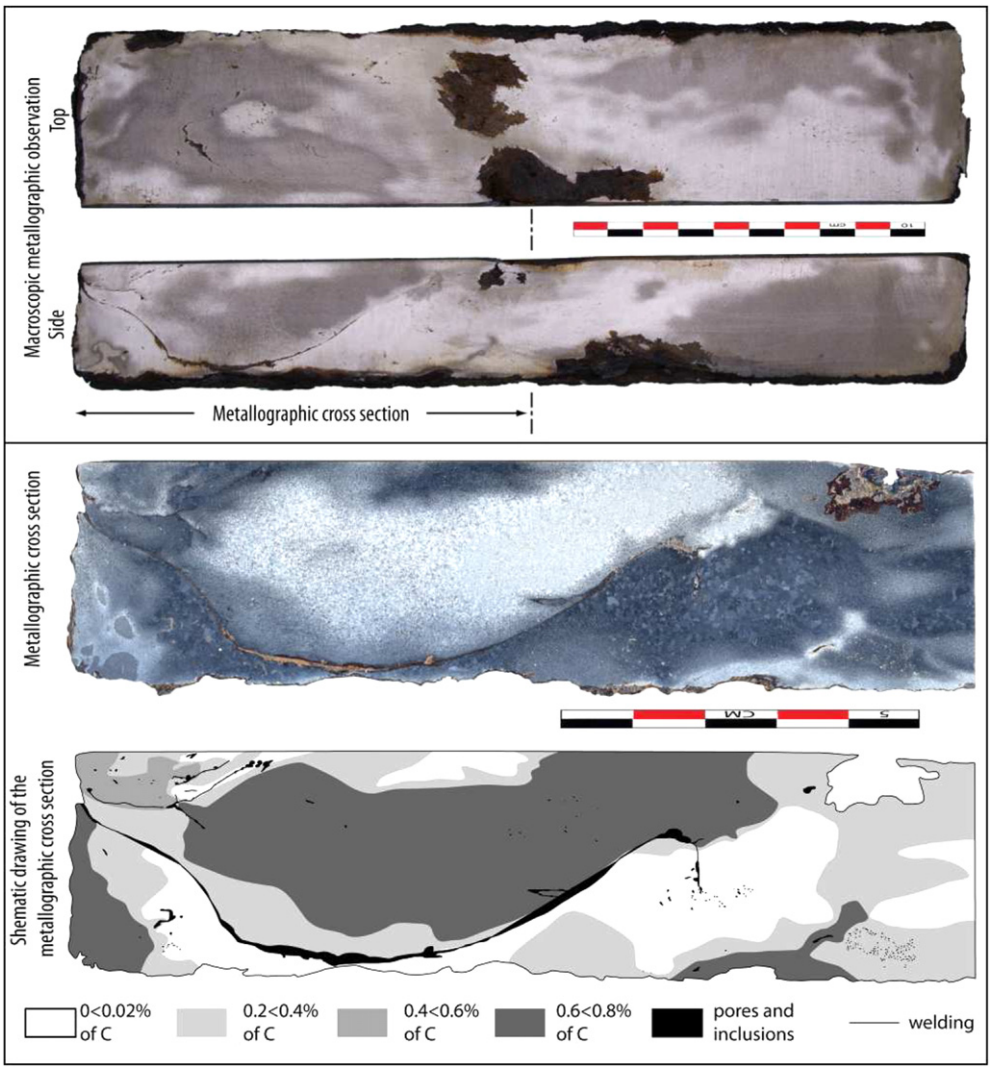

Fig. 4. An example (sample 4C SM2_5) of a microscopic metallographic observation on a bar after Nital etching with the corresponding schematic drawing of the metallographic cross section.

PPMs could be entire blooms, compacted to form a dense block of metal. Nevertheless, no unambiguous proof of this hypothesis has been published yet, and the PPMs could also have been formed from bloom fragments. Moreover, archaeological iron-based materials are generally heterogeneous, even when originating from a single smelting operation, since the process does not take place in liquid phase: ferritic iron and steel can be found together in the same bloom. Furthermore, in most cases, depending on the local thermodynamic conditions in the furnace, the steel zones are more or less carburised. In addition, the metal sometimes contains phosphorus. Finally, depending on the refining and compacting operations, a certain amount of inclusions and pores can remain in the metal. All these parameters have to be taken into account in order to evaluate the properties of the metal and its potential quality. Moreover, if an artefact is made up of several PPMs, the welding between them must also be taken into consideration.

Thus, the artefacts studied here can be characterised by six main factors: the number and the volume of the PPMs that were welded together to form the bar, the welding quality, the cleanliness of each PPM, and the carbon and phosphorus contents of the metal (Table 3).

3.1. PPM numbers and weights

Most of the bars (types 1M, 2M, 4C and 6C) are made of one PPM. Only one bar of the 2M type is made from two PPMs that are very small (Fig. 3). In contrast, types 1L (Fig. 10) and 4L (Fig. 11) are always made of several PPMs, two and four respectively (only one bar of the 4L type is made of three PPMs but is not entirely preserved). As for the 2M type, in some cases, when one of the PPM is small, three and five PPMs respectively are used.

Fig. 5 shows the weights of the PPMs for all the bars. Modal PPM weights are observed at 2 and 5 kg: 66 PPMs have a weight between 1 and 5 kg (the PPM mass is calculated by considering their proportion in the bar and the mass of the bar; Table 2, Fig. 5). These values could be explained on the one hand by the maximum quantity of metal that can be produced in a single shaft furnace, or, more probably, by the easiest workable volume by hand forging. Actually, the rare shaft furnace blooms from the Roman period found in the Alps and the Haut-Languedoc (Domergue, 1993; Fluzin, 2006) vary between 10 and 30 kg. In the bars, the heaviest PPM weight is likely around 7–8 kg. Considering this discrepancy, it is not obvious that the PPMs found in the bars are originating from

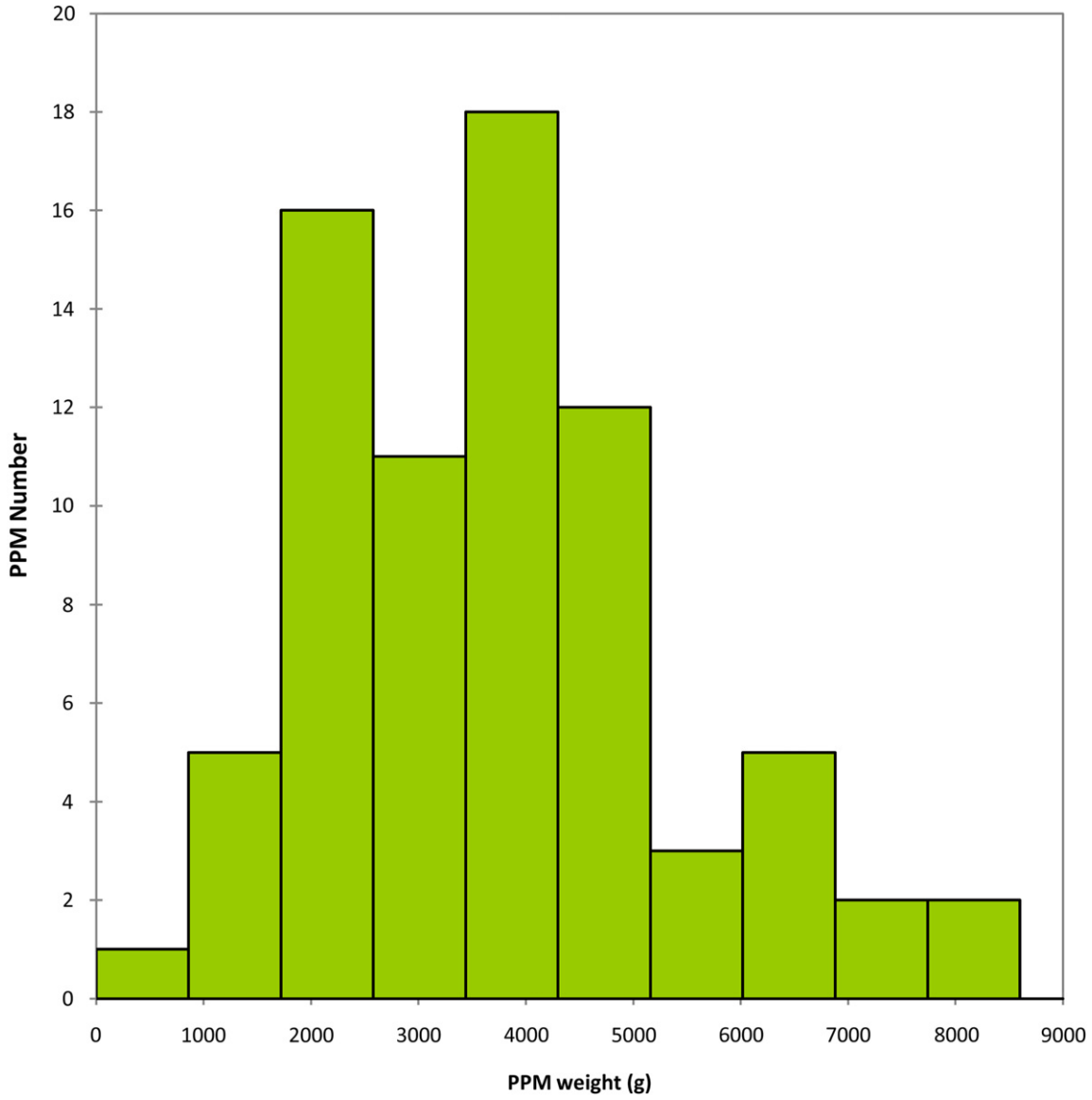

Fig. 5. Weight distribution of the PPMs for all the analysed bars.

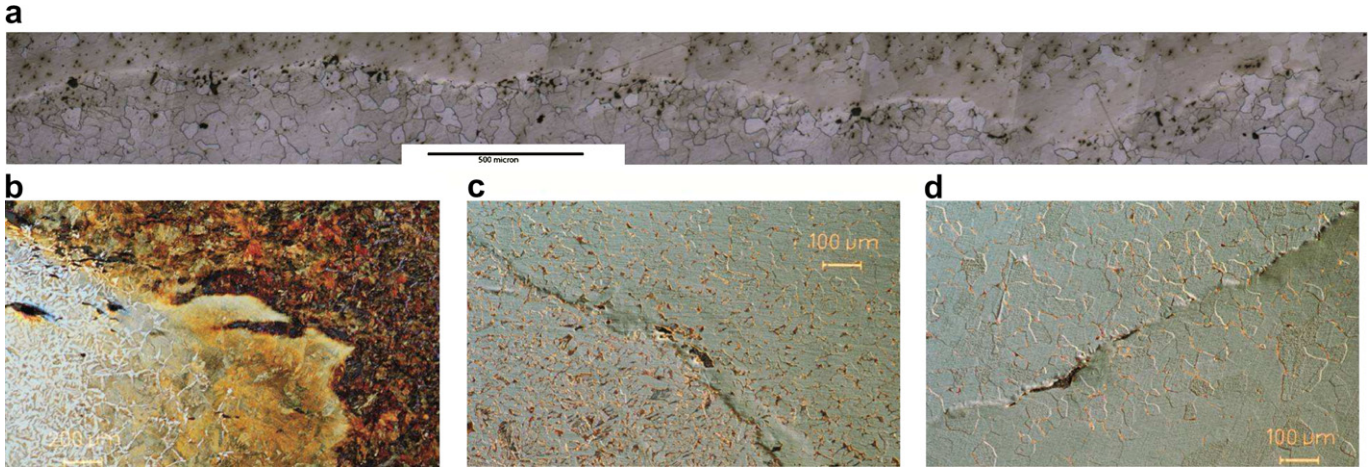

Fig. 6. Different types of welding between PPMs after Nital etching: (a) phosphorus iron–ferritic iron (4L SM24_2); (b) different steels (0.4 and 0.8% of C; 1L SM10_1), (c) steel (0.2% of C)–ferritic iron (1L SM10_2); (d) ferritic iron (1LSM10_2).

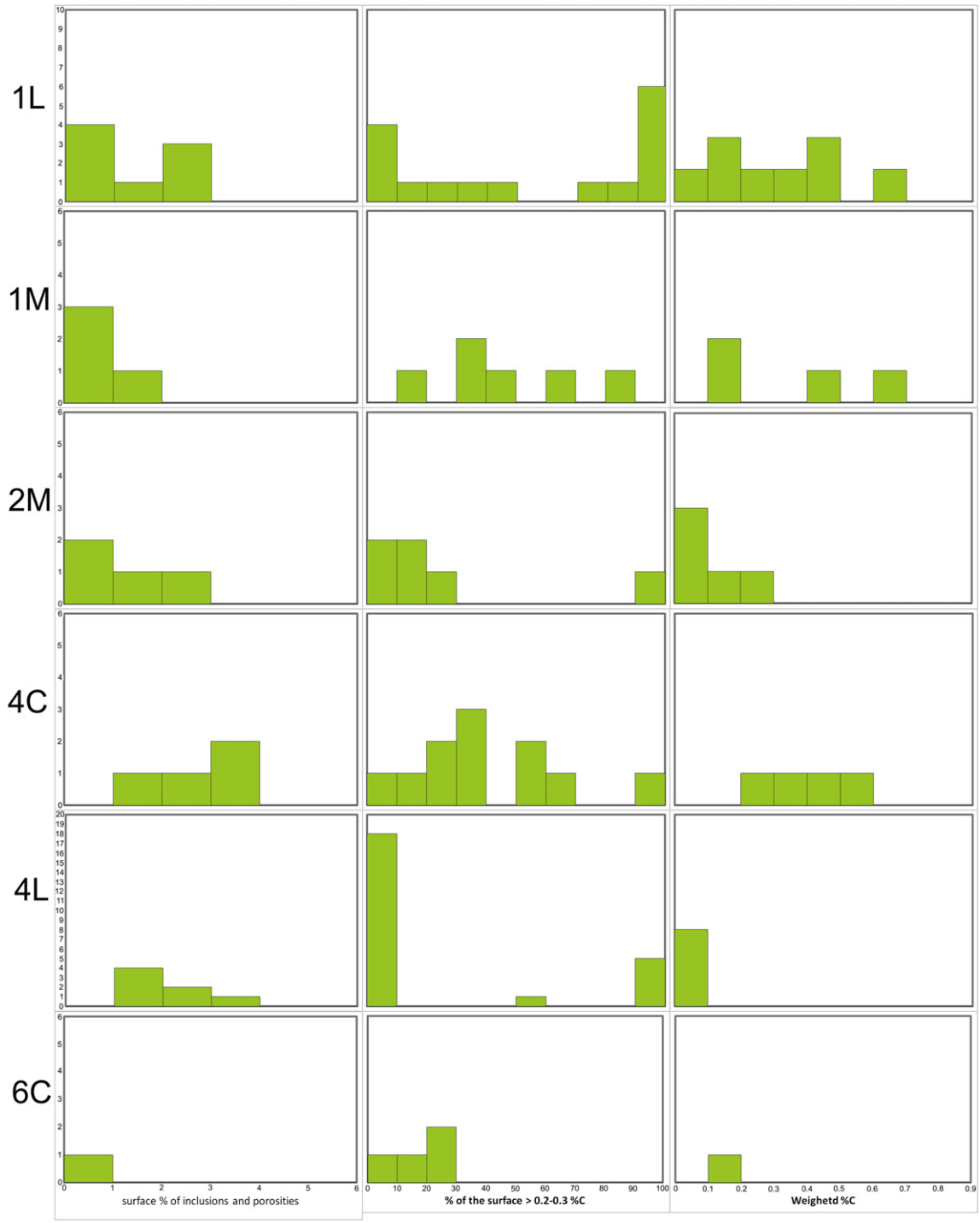

Fig. 7. Column 1: distribution of surface percentage of inclusions and pores for each PPM by type; Column 2: proportion of 'steely' surface (carbon content higher than 0.2–0.3% C) for each PPM by type; Column 3: frequency distribution of the weighted carbon contents (microscopic observations) by type.

complete compacted blooms. More probably, the initial blooms, heavier than the PPMs observed here, were first cut into smaller pieces, easier to refine and forge. This was already observed on some ancient blooms, on which traces of cutting show the intention to cut them out in several pieces (Fluzin, 2006).

3.2. Welding lines

Fig. 6 shows the different types of welding observed between PPMs. In all bars, these welding lines are carried out skillfully. Very few inclusions are visible along the welded joint. Some areas are so well welded that the welding line becomes invisible. It has to be emphasised that this type of welding, between massive pieces such as the PPMs, is particularly hard to perform in a homogeneous manner. However, whatever the morphological types (1L and 4L), the manufacture is of great quality. Nevertheless, it must be noted that, in some PPMs, welding lines formed during compacting are of significantly lower quality (see Fig. 4, for example). This difference of quality could suggest that different craftsmen performed these two operations (bloom compacting to obtain a PPM and welding between PPMs).

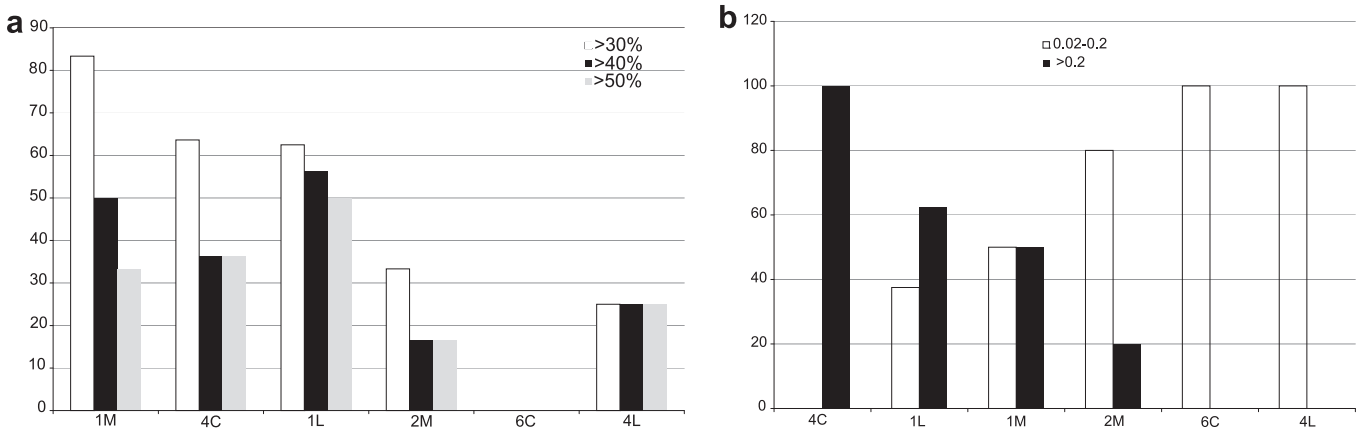

Fig. 8. (a) Percentage of the analysed PPMs with 'steely' (>0.2–0.3% C) surface (macroscopic observations) greater than 30, 40 and 50% of the total surface; (b) Percentage of the analysed PPMs with a weighted carbon content lower than 0.2 and greater than 0.2.

3.3. Metal cleanliness of the PPMs

Fig. 7 (column 1) shows the percentage of inclusions and pores for each sample. Types 1M, 2M, 6C and 1L are the cleanest (generally less than 1.5% of the observed metallographic cross sections). Types 4C and 4L present a slightly lower level of cleanliness, with an inclusion surface of up to 3.5%. Nevertheless, these values are relatively low for this type of product, as compared to other similar measurements (L'héritier and Dillmann, 2010; L'héritier et al., 2010).

3.4. Carbon and phosphorus contents of the PPM metal

Fig. 7 (column 2) displays the proportion of 'steely' surface (carbon content higher than 0.2–0.3% C) determined by macroscopic observations for all PPMs of each bar type. The values show significant variability for each type of bar: relatively carburised PPMs and also ferritic ones can always be found for the same type of bar. Nevertheless, some trends can also be distinguished. Fig. 8a shows the percentage of the PPMs having a steely surface greater than 30, 40 and 50%, respectively, of the observed surface for each type. It is then possible to distinguish two main groups. On the right side of the figure are the low carburised types, with less than 40% of the PPMs presenting more than 30% of steely surface: 2M, 6C and 4L. At the left of the figure are the most carburised types (1M, 4C and 1L). More than 60% of these PPMs have at least 30% of the surface that can be considered as 'steely'. Types 1M and 1L have 50% and more of their PPMs presenting more than 40% of 'steely' surfaces. Finally, 50% of the 1M type has more than 50% of 'steely' surfaces.

Fig. 7 (column 3) shows the weighted carbon content determined by microscopic observations for each PPM and bar type.

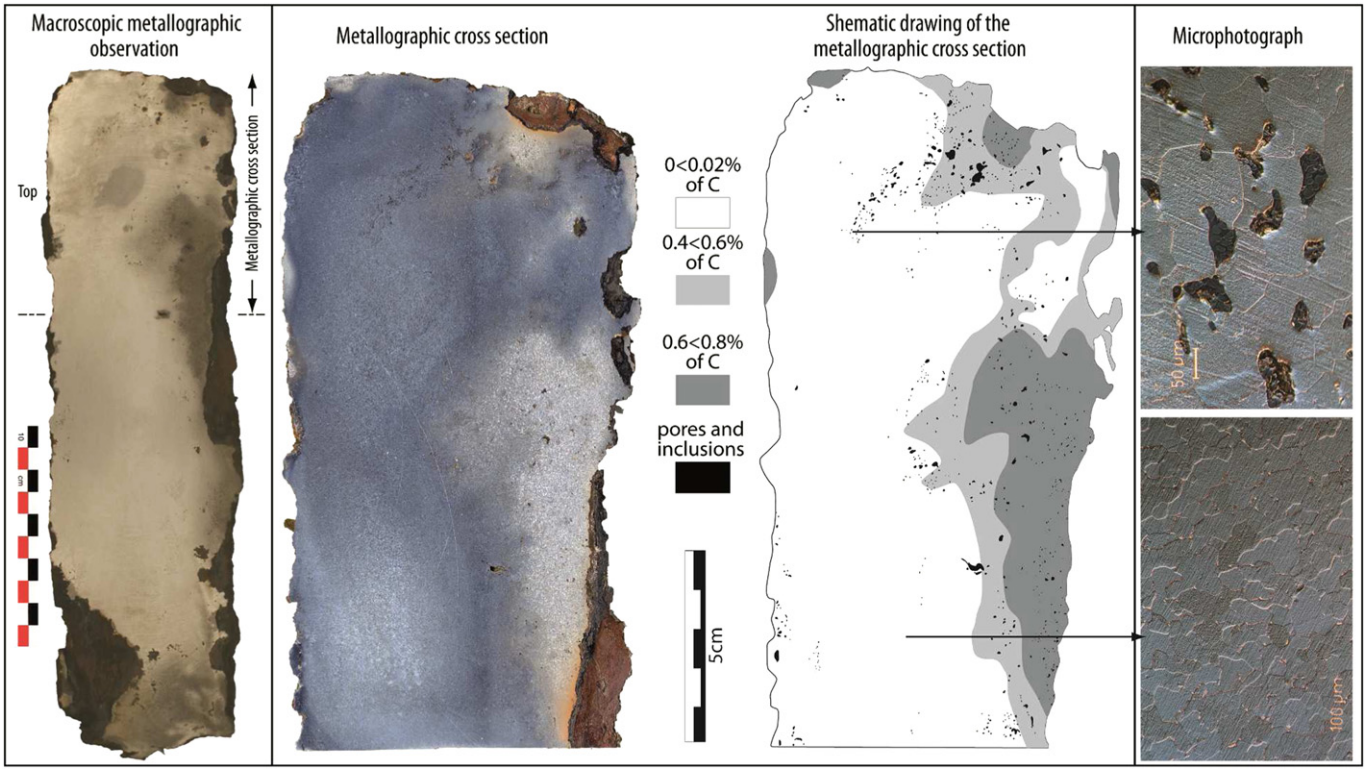

Fig. 9. An example of the metallographic observation of a ferritic bar (6C SM6_1) at the macroscopic and the microscopic scale after Nital etching.

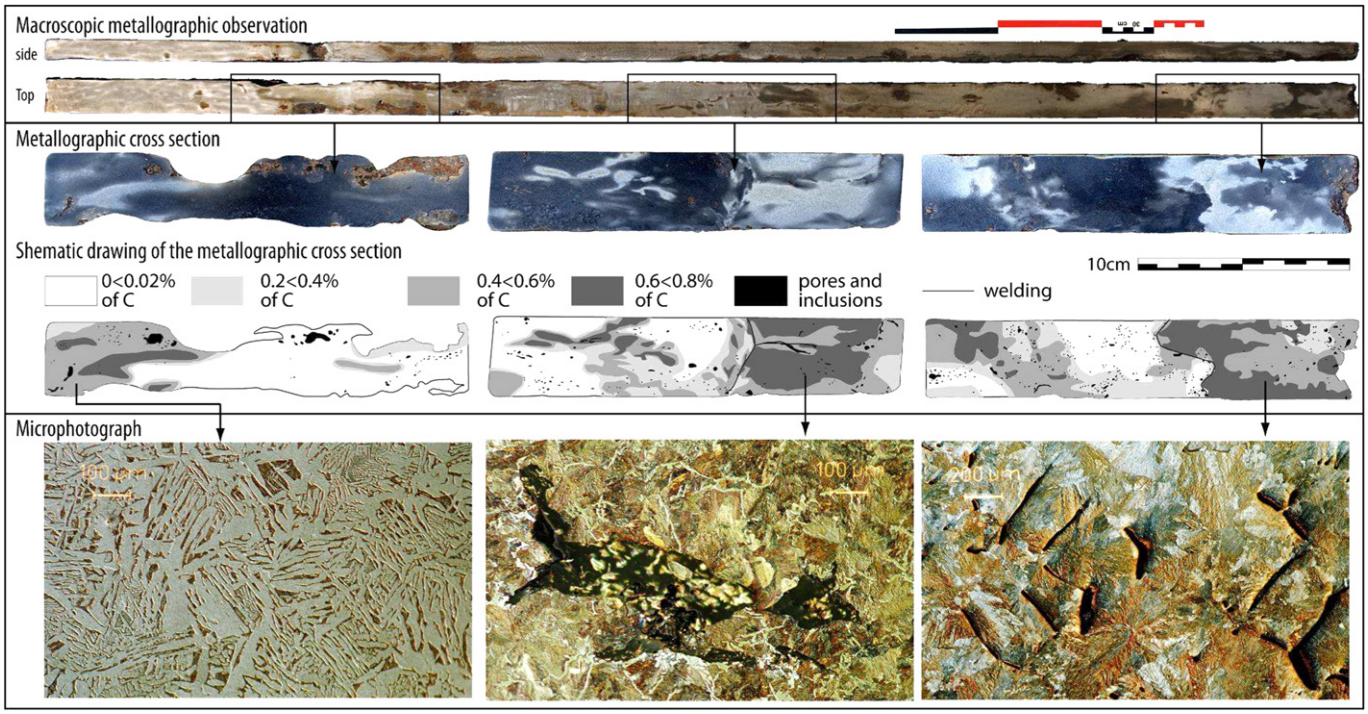

Fig. 10. An example of the metallographic observation of a steel bar (1LSM10_1) at the macroscopic and the microscopic scale after Nital etching.

Here too, despite variability, the same trend is observed: 2M, 4L and 6C types are mainly made up of low carburised alloys, more than 80% of the PPMs are made up of metal with weighted carbon contents lower than 0.2% (Fig. 8b). In contrast, the 4C, 1M and 1L types are significantly more carburised: each of these types has respectively 100%, 60% and 50% of their PPMs made of metal with a weighted carbon content over 0.2%.

Lastly, phosphorus is associated only with the 4L type. More specifically, six of the eight 4L analysed PPMs contain significant amounts of phosphorous (0.1–0.5%), also revealed by the presence of ghost structures in the ferritic structure (Fig. 11, Table 3).

Because some of the bars are made of several PPMs, it was necessary to calculate the results for entire bars. To this purpose, the weighted carbon contents and percentages of 'steely' surface

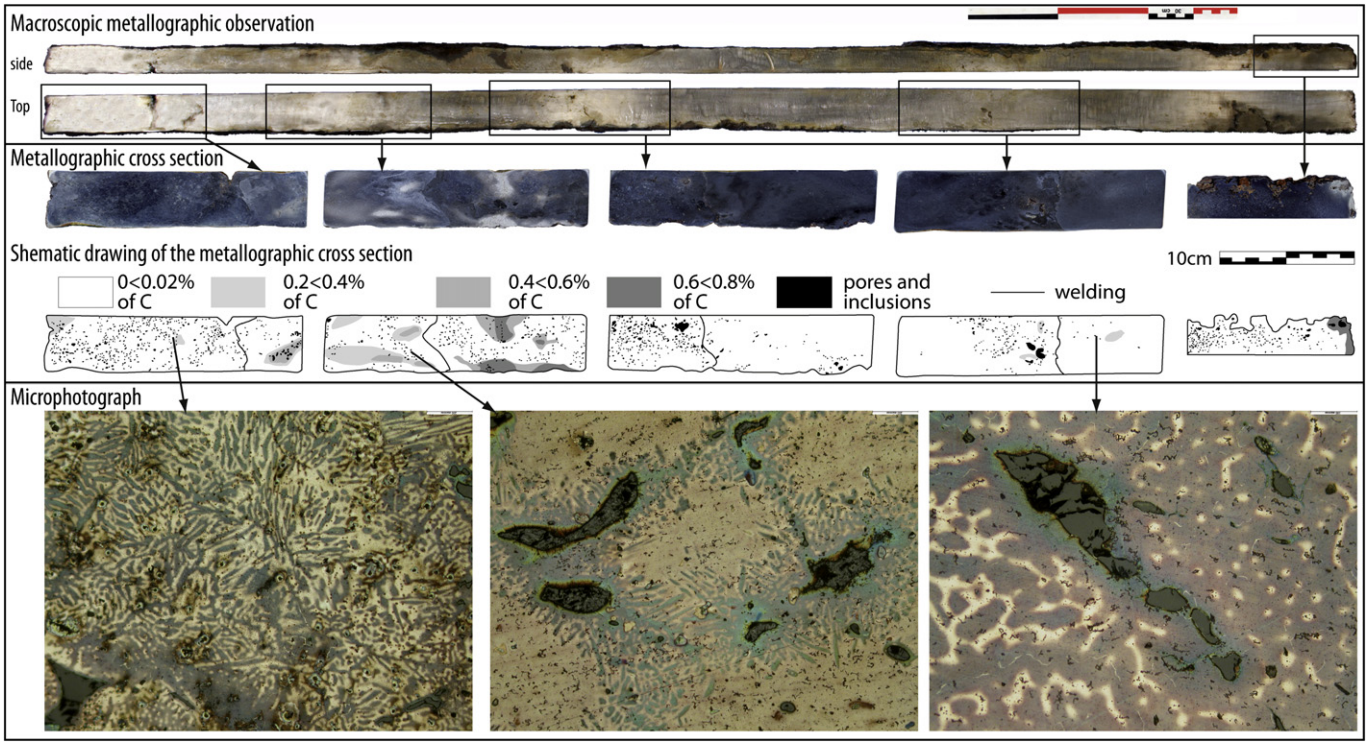

Fig. 11. An example of the metallographic observation of a phosphoric iron bar (4L SM24_2) at the macroscopic and the microscopic scale after Nital etching.

Table 3
Synthesis of the metallographic results obtained on the bars of Saintes-Maries-de-la-Mer; (MaS): macroscopic scale, (MiS): microscopic scale. Bar identification: type, wreck number, bar number, PPM number.

Bar identification	Number of PPMs	PPM weight (g)	Pores and inclusion % (MiS)	'Steely' surface %(MaS)	% of 0–0.02 wt% C area (MiS)	% of 0.02–0.2 wt% C area (MiS)	% of 0.2–0.4 wt% C area (MiS)	% 0.4–0.6 wt% C area (MiS)	%0.6–0.8 wt% C area (MiS)	%C*(MiS)	Ghost structures $0.1 < \%P < 0.6$ (MiS)
1L SM2_1 L1	3	2628	0.4	42	33	0	2	24	41	0.41	No
1L SM2_1 L2	3	507	0.5	94	1	0	7	27	65	0.61	No
1L SM2_1 L3	3	2545	0.5	19	90	8	0	0	2	0.02	No
1L SM2_2	n.d.	n.d.	0.8	n.d.	22	2	5	18	53	0.48	No
1L SM2_7 L1	2	2391	n.d.	4	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
1L SM2_7 L2	2	1492	n.d.	100	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
1L SM2_8 L1	2	2470	n.d.	100	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
1L SM2_8 L2	2	1800	n.d.	4	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
1L SM10_1 L1	2	4946	2.1	72	44	10	9	16	21	0.26	No
1L SM10_1 L2	2	4359	1.3	82	25	12	19	19	25	0.34	No
1L SM10_2 L1	2	4538	2.1	38	75	0	1	14	10	0.14	No
1L SM10_2 L2	2	3862	2.2	6	63	4	5	17	11	0.18	No
1L SM10_3 L1	3	2277	n.d.	100	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
1L SM10_3 L2	3	3916	n.d.	100	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
1L SM10_3 L3	3	3367	n.d.	10	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
1L SM10_4 L1	2	3264	n.d.	24	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
1L SM10_4 L2	2	2846	n.d.	100	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
1M SM9_5	1	3664	n.d.	16	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
1M SM9_6	1	3580	0.6	48	23	0	5	28	44	0.46	No
1M SM9_7	1	2510	n.d.	69	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
1M SM9_8	1	4000	0.6	34	76	6	3	7	8	0.11	No
1M SM9_9	1	4140	1.0	32	71	1	1	5	22	0.18	No
1M SM9_10	1	4582	0.3	82	7	0	0	0	93	0.65	No
2M SM6_3	1	3104	n.d.	13	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
2M SM6_4	1	2038	n.d.	6	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
2M SM9_1	1	3824	2.2	3	96	4	0	0	0	<0.01	No
2M SM9_2	1	2782	0.9	n.d.	100	0	0	0	0	<0.01	No
2M SM9_13	1	2310	n.d.	1	n.d.	n.d.	n.d.	n.d.	n.d.	<0.01	n.d.
2M SM25_1	1	3884	1.1	95	12	55	11	5	17	0.23	No
2M SM25_2	1	1914	0.5	14	63	0	12	10	15	0.19	No
2M SM25_3	1	4540	n.d.	18	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
2M SM25_4	1	3590	n.d.	21	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
2M SM25_5 L1	2	1053	n.d.	4	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
2M SM25_5 L2	2	921	n.d.	4	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
2M SM25_6	1	2350	n.d.	12	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4C SM2_3	1	1976	n.d.	7	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4C SM2_4	1	4258	n.d.	22	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4C SM2_5	1	3744	1.9	68	25	0	8	23	44	0.45	No
4C SM6_2	1	2838	3.6	26	53	0	8	11	28	0.28	No
4C SM6_5	1	2560	n.d.	31	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4C SM6_6	1	4240	n.d.	51	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4C SM6_7	1	3820	n.d.	54	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4C SM9_3	1	4602	3.2	36	43	0	0	0	57	0.40	No
4C SM9_4	1	3200	1.25.	n.d	83	0	0	13	4	0.1	No
4C SM9_11	1	5855	2.9	95	15	8	0	0	77	0.55	No
4C SM9_12	1	3878	n.d.	37	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4C SM24_7	1	6870	n.d.	27	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4C SM24_8	1	5130	n.d.	13	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_1 L1	4	3740	1.4	0	80	15	3	0	2	0.04	Yes. <0.5%
4L SM24_1 L2	4	3860	n.d.	2	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_1 L3	4	4690	1.6	0	93	7	0	0	0	0.01	Yes. <0.5%
4L SM24_1 L4	4	1660	1.5	6	95	5	0	0	0	0.01	Yes. <0.5%
4L SM24_2 L1	5	6280	2.8	0	94	4	2	0	0	0.01	Yes. <0.5%
4L SM24_2 L2	5	6930	1.3	1	80	7	5	3	5	0.07	Yes. <0.5%
4L SM24_2 L3	5	4280	2.5	0	78	8	5	3	6	0.08	No
4L SM24_2 L4	5	3130	1.3	0	99	1	0	0	0	<0.01	No

4L SM24_2 L5	5	2270	3.4	3	92	0	0	8	0.06	Yes, <0.5%
4L SM24_3 L1	4	8590	n.d.	0	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_3 L2	4	6768	n.d.	0	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_3 L3	4	5114	n.d.	0	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_3 L4	4	6668	n.d.	0	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_4 L1	3	4895	n.d.	57	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_4 L2	3	5448	n.d.	92	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_4 L3	3	7527	n.d.	2	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_5 L1	4	3768	n.d.	0	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_5 L2	4	2327	n.d.	0	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_5 L3	4	2080	n.d.	2	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_5 L4	4	5745	n.d.	1	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_6 L1	4	1634	n.d.	94	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_6 L2	4	2653	n.d.	95	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_6 L3	4	3428	n.d.	97	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
4L SM24_6 L4	4	1765	n.d.	98	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
6C SM6_1	1	4910	0.5	25	71	0	6	9	0.16	No
6C SM6_8	1	8120	n.d.	19	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
6C SM6_9	1	4890	n.d.	27	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
6C SM6_10	1	6790	n.d.	7	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.

were calculated for each bar, taking into account the representativeness of each PPM (i.e. the observed surface of each PPM). For P content, because no numerical value was measured, the bar was considered as made of P-iron if more than 50% of its PPMs showed ghost structures (Table 4).

The same trends as for individual PPMs can be observed. All the bars of the 1L type have more than 20% of steely surface and more than half of them have over 50% of steely surface, with some of the bars reaching 90%. Unfortunately, only three bars were studied at the microscopic scale. For these bars, weighted carbon contents are comprised between 0.1 and 0.3%. Considering these values, it can be deduced that the bars belonging to the 1L type observed in this study are relatively carburised, and with a good cleanliness.

2M and 4L bars present slightly different features. The only 2M bar made of 2 PPMs has less than 5% of steely surface. The same can be said for the 4L type: four of the six analysed bars have a steely surface lower than 10%. The weighted carbon contents of the two bars analysed at the microscopic level are under 0.1%. Moreover, this type is the only one made of phosphoric iron. To sum up, the 2M and 4L bar types analysed here seem mainly to belong to the category of ferritic iron materials. The difference between them is linked to the phosphorus content.

3.5. Nature and quality of materials

Considering all the bars analysed in the present study, it seems that the different types could be classified into several categories of materials. Types 1L and 1M consist of bars mainly made of steel (Fig. 10). Types 6C, 2M and 4L contain mainly specimens of ferritic iron (Figs. 3 and 9); moreover, type 4L is made of phosphoric iron (Fig. 11). The 4C type cannot be classified as it consists of various products, without any clear trend, and may be considered as heterogeneous (Fig. 4).

As far as cleanliness is considered, no difference between the different types of bars was observed. All specimens showed very good cleanliness. Nevertheless, the number of PPM welding junctions differentiate the bar types. It is obvious that the types of bars consisting of numerous PPMs (types 1L and 4L) required significantly more work for their manufacture. Consequently, these different types required different types and amounts of work for their production, and should have had different commercial values. Moreover, the presence of welding lines would have probably induced local weaknesses in the bar that could have also influenced its commercial value. Lastly, these differences can also be explained by the different shapes of the products. Thus, 1L and 4L bars, with

Table 4
Metallographic characteristics of the bars made of more than one PPM. Average contents weighted by the surface of each constitutive PPM. Bar identification: type, wreck number, bar number, PPM number.

Bar identification	'Steely' surface %	Pores and inclusions %	(%C*)	P-iron
1L SM2_1	36	0.5	0.25	No
1L SM2_7	41	n.d.	n.d.	n.d.
1L SM2_8	60	n.d.	n.d.	n.d.
1L SM10_1	77	1.7	0.30	No
1L SM10_2	23	2.1	0.16	No
1L SM10_3	68	n.d.	n.d.	n.d.
1L SM10_4	59	n.d.	n.d.	n.d.
2M SM25_5	4	n.d.	n.d.	n.d.
4L SM24_1	1	0.9	0.02	Yes
4L SM24_2	1	2.1	0.05	Yes
4L SM24_3	0	n.d.	n.d.	n.d.
4L SM24_4	45	n.d.	n.d.	n.d.
4L SM24_5	1	n.d.	n.d.	n.d.
4L SM24_6	96	n.d.	n.d.	n.d.

Table 5
* Slag inclusion composition for detected compounds (weighted content) for each analysed bloom. Normalised to 100. Na, S, Cl, Ti, V, Cr oxides were quantified but not presented in the table because their respective contents are always lower than 0.5%. ** Contents normalised at 100% without FeO. Bar identification: type, wreck number, bar number, PPM number.

Bar PPM	MgO*	Al_2O_3*	SiO_2*	P_2O_5*	K_2O*	CaO*	MnO*	FeO*	P_2O_5**	MnO**
1L SM2_1 L1	0.2	2.9	15.6	1.5	1.4	1.8	1.1	74.2	5.9	4.1
1L SM2_1 L2	2.2	11.5	44.0	1.0	3.3	4.1	0.8	31.9	1.5	1.2
1L SM2_1 L3	2.4	13.2	54.4	0.5	4.3	5.8	1.0	17.5	0.6	1.2
1L SM2_2	2.9	14.3	58.7	0.6	3.4	4.3	1.6	13.1	0.7	1.8
1L SM10_1 L1	2.9	6.7	33.6	0.4	2.1	4.1	1.5	47.7	0.8	2.8
1L SM10_1 L2	3.2	8.9	42.7	0.3	3.0	4.3	1.4	34.9	0.5	2.1
1L SM10_2 L1	1.2	4.3	22.9	0.4	1.5	2.4	0.9	65.8	1.0	2.5
1L SM10_2 L2	3.0	8.3	40.7	0.2	2.7	3.1	1.0	39.6	0.4	1.6
1M SM9_6	1.8	10.6	32.9	0.3	2.3	3.3	2.1	45.6	0.9	1.4
1M SM9_8	0.3	4.8	25.7	0.3	1.1	0.8	0.5	65.8	2.3	1.2
1M SM9_9	0.3	3.4	18.8	0.7	1.2	2.4	0.4	72.0	0.3	1.4
1M SM9_10	3.6	12.3	49.4	0.3	4.0	6.9	0.7	21.2	0.3	0.9
2M SM9_1	0.2	3.6	23.2	2.2	1.2	1.9	1.6	65.2	1.8	0.5
2M SM9_2	0.4	4.1	34.4	0.8	1.7	2.6	0.2	54.6	0.4	2.0
2M SM25_1	1.5	11.6	52.3	0.3	2.4	4.2	1.0	25.2	1.9	2.7
2M SM25_2	0.8	6.5	32.6	0.9	1.8	2.7	1.3	52.2	6.4	4.6
4C SM2_5	1.7	8.9	47.2	0.3	4.1	9.3	1.5	25.7	0.4	1.5
4C SM6_2	0.5	4.8	31.1	0.3	2.0	3.4	3.4	53.8	1.0	0.7
4C SM9_3	2.1	11.6	43.8	0.6	3.1	2.5	0.5	34.9	14.4	0.8
4C SM9_4	1.4	8.1	40.1	1.2	2.3	1.9	2.2	41.7	21.9	1.5
4C SM9_11	3.5	13.3	51.6	0.3	3.1	5.3	1.2	19.6	32.8	0.8
4L SM24_1 L1	0.2	3.0	15.3	10.8	0.8	1.6	0.3	67.3	2.8	0.6
4L SM24_1 L3	0.9	6.9	29.9	7.4	2.2	2.8	0.4	48.7	1.7	6.3
4L SM24_1 L4	0.9	5.5	30.8	8.6	2.1	5.0	1.3	44.7	1.5	6.4
4L SM24_2 L1	0.3	3.6	21.1	8.8	1.3	2.9	0.6	60.9	3.1	3.2
4L SM24_2 L2	0.6	5.8	26.6	1.1	1.6	3.0	0.3	60.2	0.5	7.2
4L SM24_2 L3	0.3	3.1	25.9	0.6	1.3	2.0	2.3	63.6	1.2	5.6
4L SM24_2 L4	0.6	4.1	38.2	0.8	1.7	1.7	3.3	49.0	15.6	2.4
4L SM24_2 L5	1.2	9.2	47.7	2.1	2.1	2.0	2.2	32.5	2.1	3.8
6C SM6_1	1.2	5.5	29.7	0.6	1.7	4.8	2.6	53.0	0.5	3.9

a very long shape, would have been used for different purposes than the other types.

3.6. Slag inclusion analyses

As for metallographic investigations, it is not the bar types that need to be differentiated by slag inclusion studies, but the PPMs. This aspect is crucial because, as demonstrated before, types 1L and 4L bars are made of more than one PPM, which could have different origins. Table 5 sums up the weighted compositions for the detected elements in each analysed PPM.

In a first step, we considered P_2O_5** and MnO** weighted amounts normalised to 100 without FeO. Except in ore, MnO is not present in significant quantities in the other components of the reduction system. Thus, if high levels of manganese are detected in the inclusions, the artefact was probably produced from a manganese ore. It has to be noted that, even for Mn-rich ores, variations of their contents at the decimetric level are possible. Nevertheless, recent studies (Leroy, 2010) on slag inclusions clearly showed that, for example when manganese-rich ores are used (1–10 wt%), the lowest content observed in the slag inclusions of the obtained metal is higher than 2 wt% and can reach 20 wt%.

P_2O_5 can only be present in significant quantities in the ore and, to a lower extent, in the charcoal. Moreover, during the reduction, a certain quantity of P_2O_5 is reduced and diffuses into the metal. Nevertheless, previous studies (Vega et al., 2002, 2003) demonstrated that very high quantities of P_2O_5 in the slag inclusions are linked to an initial presence in the ore. Thus, in this case as well, specimens containing slag inclusions with high levels of P_2O_5 can be distinguished from the rest of the artefacts.

Fig. 12 shows the P_2O_5** and MnO** weighted contents for the different samples. It appears clearly that at least three different categories can be distinguished on the diagram on these bases. Samples 4L SM24_1 L4, 4L SM24_2 L1, 4L SM24_1 L3 and 4L SM24_1 L1 all present weighted phosphorus contents (normalised at 100) higher than 6%, linked with the presence of phosphorus in the metallic matrix (Table 3). These high amounts can probably be explained by the use of a phosphoric ore and reveal a different origin than the other PPMs. It has to be emphasised that PPMs with relatively high phosphorus slag inclusions do not necessarily originate from the same place, because P-ores are frequent.

Another sample category can be distinguished by relatively high MnO** contents, i.e. over 5% (samples 4C SM6_2, 4L SM24_2 L4, 4L SM24_2 L3 and 6C SM6_1). These contents also reveal different origins from the first category (high P_2O_5** amounts), as well as the rest of the samples showing lower amounts of MnO**. Once again, these high amounts of MnO in the slag inclusions are not proof of a common origin. For example, two of these samples (4L SM24_2 L4 and 4L SM24_2 L3) have the lowest Al_2O_3*/SiO_2* ratios, showing a different reduction system from the other high MnO** samples (Fig. 13), as well as the rest of the samples.

For some other samples (2M SM9_1, 1M SM9_6, 1L SM2_1 L1, and 4L SM24_2 L5), average quantities of MnO** were also detected and are comprised between 3 and 5 wt%. These samples originate from a different reduction system than the ones with a low MnO** content. It has to be noted that 4C SM9_4 has an average MnO** content, but also presents a higher K_2O*/CaO* ratio, and can thus be differentiated.

Fig. 14 shows the NRC ratios calculated on the weighted composition of slag inclusions. As for MnO** and P_2O_5**, different groups can be distinguished. One group, consisting of samples 1L SM10_1 L1, 1L SM10_1 L2 and 1L SM10_2 L2, can be characterised by a significantly higher MgO*/Al_2O_3* ratio than the other samples. It is interesting to note that these samples do not have either high P_2O_5** contents, or high MnO** contents, and consequently they originate from different reduction systems than the previous ones.

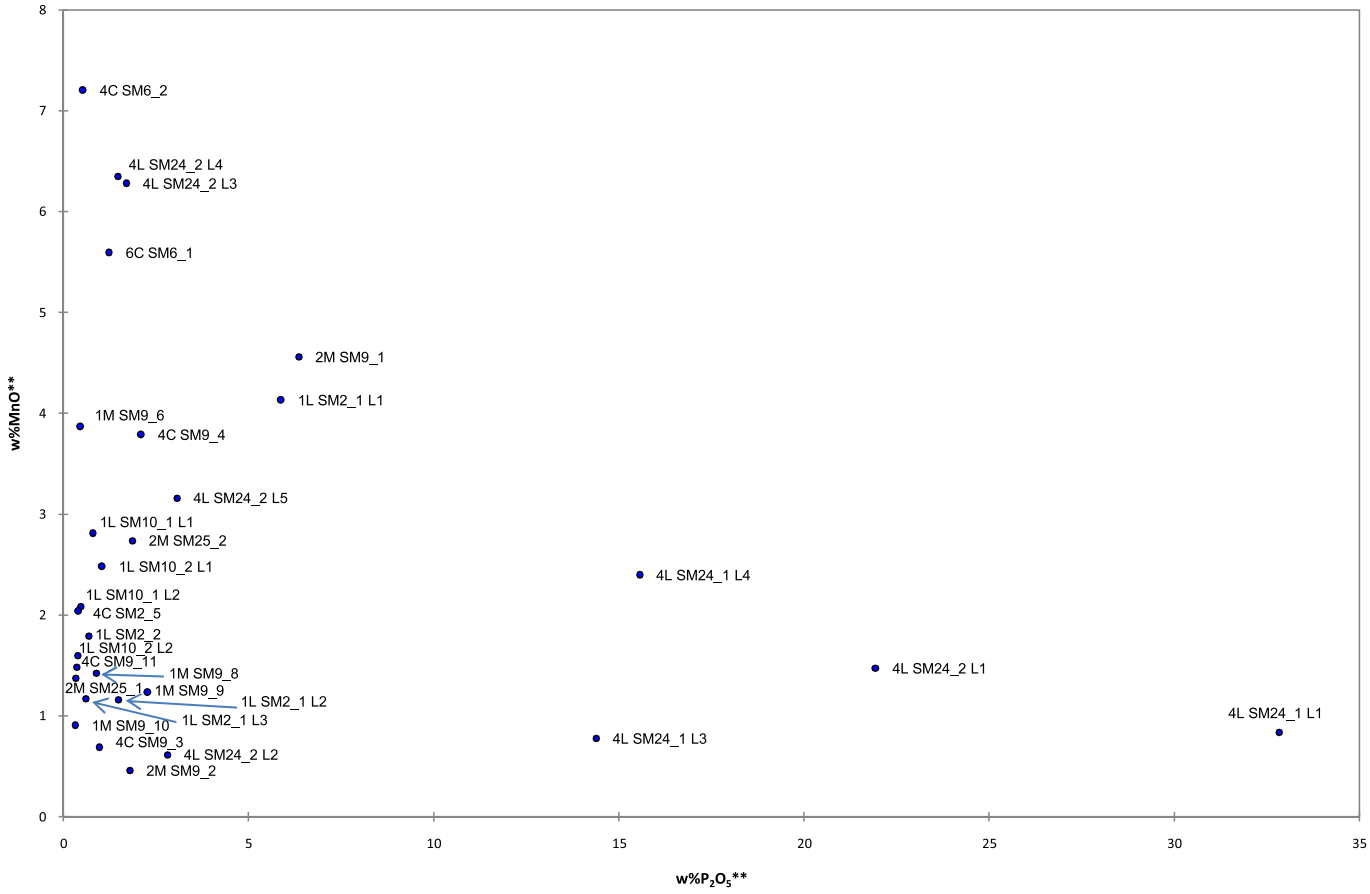

Fig. 12. P_2O_5** and MnO** contents measured in the inclusions of the different samples.

If we are to look more carefully at all these compositional data, other groups can be distinguished. Three samples present a low Al_2O_3*/SiO_2* ratio (Fig. 13) and are consequently originating from different reduction systems than all the other samples. As it has been said before, two of them (4L SM24_2 L4 and 4L SM24_2 L3) also have a high MnO** content and thus originate from a different reduction system than the last one (2M SM9_2).

Two other samples (4C SM9_3 and 1M SM9_8) have a higher K_2O*/CaO* ratio and low P_2O_5** and MnO** contents. Consequently, they originate from different reducing systems. These two samples could probably also be differentiated by their Al_2O_3*/SiO_2* ratio.

This short comparison of the slag inclusion analytical data clearly indicates that the different PPMs were produced in various reduction systems. Major element analyses alone do not allow validating hypotheses of common origins, because the same NRC ratios could correspond to several reduction systems. Nevertheless, the difference between NRC ratios, MnO** and P_2O_5** contents clearly validate the hypothesis of different origins. Consequently, we decided to group the samples in different categories, linked to these parameters. It has to be emphasised that these categories only indicate differences between samples. It is not because two artefacts belong to the same category that they have the same origin. In order to be able to provide an answer to that question, it would be necessary to perform trace element analyses.

At least six different reducing systems categories can be distinguished (Table 6):

- Category 1: High MnO** contents, the NRC ratios belong to the main common group observed in Figs. 13 and 14.
  Category 1a: High MnO** contents, lower Al_2O_3*/SiO_2* ratios than the main group in Figs. 13 and 14.
  Category 1b: Average MnO** contents, the NRC ratios belong to the main common group observed in Figs. 13 and 14.
  Category 1c: Average MnO** contents, higher K_2O*/CaO* NRC ratios.
- Category 2: High P_2O_5** content.
- Category 3: Higher MgO*/Al_2O_3* ratios without significant MnO** or P_2O_5** levels.
- Category 4: Lower Al_2O_3*/SiO_2* ratios without significant MnO** or P_2O_5** levels.
- Category 5: Higher K_2O*/CaO* without significant MnO** or P_2O_5** levels.
- Category 6: no particular composition (main group in Figs. 13 and 14, no significant amounts of P_2O_5** and MnO**).

Considering these results, a single reduction workshop cannot be exclusively linked to a particular type of bar. Moreover, the fact that some bars (for example 1L SM10_2 or 4L SM24_2) are made up of PPMs with different chemical signatures of major elements and NRC ratios (originating from different workshops) could indicate that bars were not systematically forged at the reduction site, as it was often asserted for this period. Also, it can be proposed that bars were made in different workshops, far from the bloomery site. These manufacturing sites probably imported products from different workshops to manufacture the bars. Nevertheless, it has to be noted that some bars (1L SM10_1) could have been made with PPMs from the same workshop, belonging to the same category. Last but not least, no indication about that issue can be inferred from the analysis of the bars constituted of a single PPM.

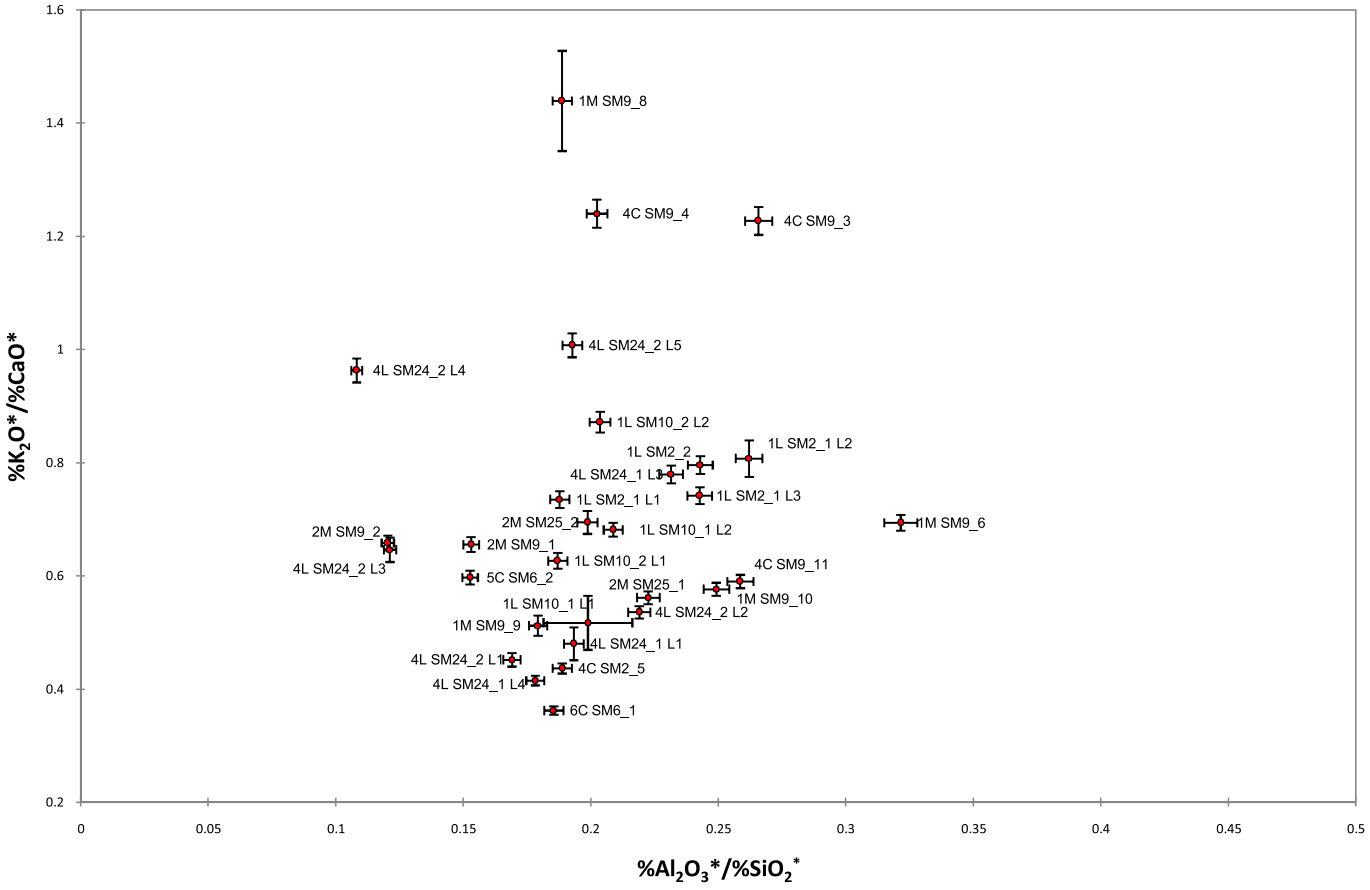

Fig. 13. NRC ratios in the slag inclusions: $Al_2O_3^*/SiO_2^*$ versus K_2O^*/CaO^*.

It is also important to emphasise that the different PPM categories are not exclusively associated to given morphological types of bars, as it seems to be the case for metal quality. Consequently, there is no clear link between the PPM type and the morphological type. It is also interesting to note that each wreck can contain different categories of PPMs (i.e. resulting from different reduction systems), indicating that their loads probably did not originate from the same workshop and were not consisting of a single type of bar. To sum up, each ship could transport different types of bars made from PPMs from various reduction systems.

4. Discussion

4.1. Existence of bar manufactures

At present, the major element study of the slag inclusions entrapped in the bars of the Saintes-Maries-de-la-Mer wrecks reveals at least nine chemical categories distinguishing various reduction systems and probably workshops (provenances). Once again, it needs to be emphasised that two PPMs belonging to the same group could also have a different origin. Major element analysis does not allow taking this discrimination further. Moreover, among the six bar types examined here, the three made of numerous PPMs seem to have been made with metal pieces from at least three reduction systems. These observations are not compatible with the hypothesis, that the workshops where the bars were manufactured were at the same location as the smelting sites where the iron of the PPMs was obtained from the ore (complete iron making *chaîne opératoire* at the same geographical site). In contrast, our observations strongly suggest a subdivision of the production line into several sectors. This subdivision was also observed in other 'proto-industrial' (indicating the massive and concentrated character of the productions) production organisations, including those of the early Roman Empire: *amphorae*, *terra sigillata*, wine, etc. (Laubenheimer, 1990, 2001).

The specialisation of the different stages of the iron making production line could also be detected by the use of standards (forms that are linked to specific metal qualities), but is also revealed by the technical quality of the forging operations revealing a specific skill (welding between PPMs, refining and compacting of the metal, especially for particular types such as 1M, 1L and 6C). In fact, despite the numerous iron ore smelting sites discovered by archaeologists (Sablayrolles, 1989; Decombeix et al., 2000; Izard and Mut, 2007; Mut and Kotarba, 2007), no data is available either regarding workshops where PPMs and bars were manufactured, or about their geographical relationships with the primary smelting sites and/or redistribution centres. These workshops could have been located near the smelting sites, but also near harbours, such as Narbonne, which is considered by Strabo as the first emporium of Gaul. Nothing allows us to determine if these workshops were grouped or dispersed; situations could be diverse. Nevertheless, in the case examined here, the fact that some bars are constituted of PPMs from different reduction systems seems to show that these manufacturing workshops were located relatively far from the smelting centres, or at least at equal distance from different centres.

Finally, we can put forth the hypothesis that, upstream, ore was smelted in regional 'proto-industrial' centres that exploited specific deposits. Subsequently, the resulting blooms, probably after a first preliminary refining that took place near the smelting site, as attested by archaeological finds (Rebiscoul, 2003; Fabre, 2004),

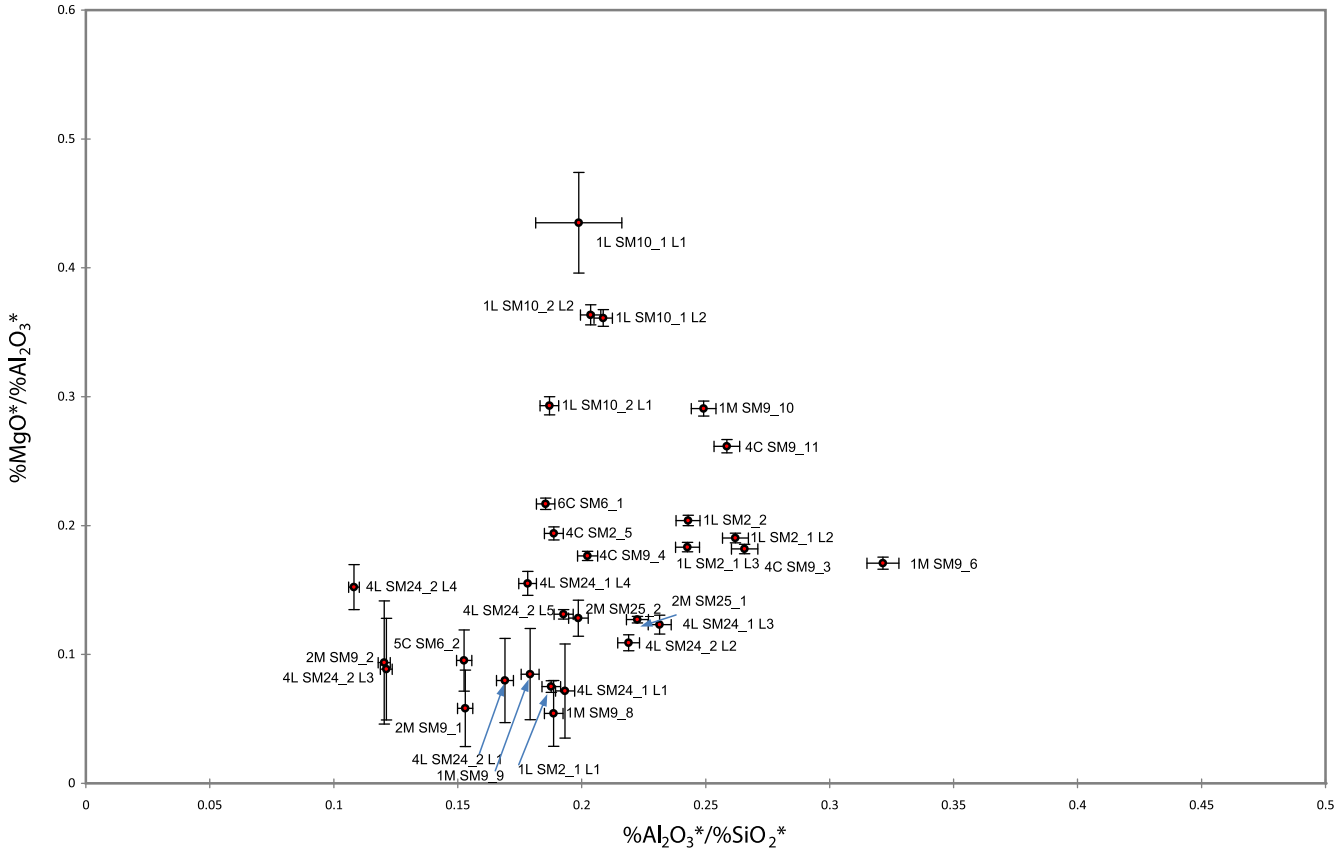

Fig. 14. NRC ratios in the slag inclusions: AL_2O_3*/SIO_2* versus $MgO*/Al_2O_3*$.

Table 6
PPM categories.

Sample	Type	Wreck	Category
1L SM2_1 L1	1L	SM2	1b
1L SM2_1 L2	1L	SM2	6
1L SM2_1 L3	1L	SM2	6
1L SM2_2	1L	SM2	6
1L SM10_1 L1	1L	SM10	3
1L SM10_1 L2	1L	SM10	3
1L SM10_2 L1	1L	SM10	6
1L SM10_2 L2	1L	SM10	3
1M SM9_8	1M	SM9	5
1M SM9_9	1M	SM9	6
1M SM9_6	1M	SM9	1b
1M SM9_10	1M	SM9	6
2M SM9_1	2M	SM9	1b
2M SM9_2	2M	SM9	4
2M SM25_1	2M	SM25	6
2M SM25_2	2M	SM25	6
4C SM2_5	4C	SM2	6
4C SM6_2	4C	SM6	1
4C SM9_3	4C	SM9	5
4C SM9_4	4C	SM9	1c
4C SM9_11	4C	SM9	6
4L SM24_1 L1	4L	SM24	2
4L SM24_1 L3	4L	SM24	2
4L SM24_1 L4	4L	SM24	2
4L SM24_2 L1	4L	SM24	2
4L SM24_2 L2	4L	SM24	6
4L SM24_2 L3	4L	SM24	1a
4L SM24_2 L4	4L	SM24	1a
4L SM24_2 L5	4L	SM24	1b
6C SM6_1	6C	SM6	1

were exported to workshops specialised in bar manufacturing. Moreover, despite variability, general trends observed in the metallographic results seem to indicate that, depending on the metal quality (ferritic iron, steel or phosphoric iron), craftsmen forged the bar under a specific form that could define a standard. Further studies must be conducted to determine if this configuration corresponds to a mainstream tendency during that period or not.

No multi-PPMs bar types could be linked to a single smelting system, i.e. a unique supply place. Most smelting places thus seem to have produced blooms of variable quality, which in turn seem to have been subsequently evaluated downstream, in the bar workshops. Then PPMs were probably forged and welded together to obtain bars with a morphology that would correspond to their properties. It can be argued that the remarkable quality of some welding (types 1L and 4L, for example) could represent the 'signature' of specific workshops with particularly skilled craftsmen. Furthermore, considering the weights of the PPMs – indicating that they could originate from selected parts of a cut bloom, a practice attested for other periods (Verna, 2001) – and the global good quality of the work (compared to the same type of bar for other periods or places), the blacksmiths seemed to have completely mastered their work, probably through precise organisation (task synchronising, adapted infrastructures, etc.) (Pagès et al., 2008, 2011; Pagès, 2010).

4.2. A standard corresponding to the quality of iron?

The forms of the bars found in the wrecks of the Saintes-Maries-de-La-Mer suggest the occurrence of a morphological standardisation at the beginning of the Roman period in the north-

western Mediterranean area. In order to explain the significance of these standardisations, several hypotheses were proposed: mass and volume of materials linked with fiscal laws governing the trade, provenance of materials (Domergue and Liou, 1997; Coustures et al., 2003, 2006). The present work, considering that the forms of the bars are indicative of trends in their metallographic structure (carbon and phosphorus contents, refining degree), proposes that the bar standardisation could also have been linked to the quality of the metal.

Three hypotheses can then be put forth: iron ore smelters, blacksmiths who forged the bars, and traders knew how to recognise ferritic iron, steel, or phosphoric iron. They also knew how to differentiate the refining and compacting degree within each of these categories. As a result, they were keenly aware of the nature of the product and had a feeling of its 'mechanical properties' (as they are referred to since the end of the 19th century). These mechanical properties were perceptible for the blacksmith through impressions of the material under the hammer or its capacity to be deformed, as it is still the case for blacksmiths nowadays (Merluzzo and Forrières, 1990; Coulibaly et al., 1999). Moreover, Pliny the Elder mentions in his *Naturalis Historia* (XXXIV) different qualities of iron metal: soft iron that can be associated to ferritic structures, and brittle iron, perhaps more comparable to steel or phosphorus iron.

The existence of a normalisation implies that it was accepted by producers, traders, and final users. The homogeneity of this standard suggests that it was accepted by numerous production and importing sites. Consequently, one of the future challenges, in addition to confirming these trends by analysing more bars, will be to delimit the geographical and chronological extension of the standard. Type 2M is for example also attested later in the 3rd century AD in the Saone region or near Vichy (Allier-France) (Bonnamour, 2000a,b; Pagès, in press), and the first metallographic analyses clearly demonstrate that the metallographic structure corresponds to a ferritic alloy, as in the present study. It could also be assumed that the regions of North Gaul, producing large quantities of iron (Dunikowski and Cabboi, 1995; Rebiscoul, 2003), also provided blooms or PPMs that were used to obtain bars, following the standard identified for the Saintes-Maries-de-la-Mer bars. In the future, it will thus be crucial to analyse further bars and half-products of the same period, following the same methodology, in order to verify these assertions.

5. Conclusion

This study on bars from the Roman Empire shows the necessity of examining on the one hand a large number of samples, and on the other hand of performing representative sampling in order to assess global trends linked to metal quality and nature. These assertions are also true concerning the distinction between different reduction systems according to major element slag inclusion analyses. Consequently, a methodology was proposed in this paper coupling observation at macroscopic and microscopic scales with relatively 'classic' and easy to perform analytical methods. Despite the significant number of bars and samples analysed here, the number of results is not sufficient to carry out statistical analyses, because of the large number of parameters that have to be considered to understand the quality of a metal produced by the bloomery process. Nevertheless, the results allow us to observe some trends in the metallographic structures and slag inclusion compositions that lead to distinguishing different types and qualities of metal that can be compared to the archaeological data. Consequently, this study cannot of course identify the exact origin of the bars, but shows the complexity of the research pertaining to the chemical signature of a significant corpus of bars. Indeed, this complexity is linked on the one hand to the fact that different reduction sites could be linked to the production of the blooms and PPMs making up the products, and on the other hand to the fact that the bar manufacturing sites do not seem to be automatically linked to the ore smelting sites.

Nevertheless, the present results highlight several aspects. During the early Roman period, it seems that at least part of the primary production of iron was dispatched to one or several cleansing workshops located far from the ore smelting site; in turn, such workshops seem to have centralised this production towards the manufacture of bars intended for export.

As far as typology is concerned, the notion of the standardisation of bars has been discussed for several years. This discussion is now open for other products as well, such as *amphorae*. To date, the standard was essentially associated to origin or weight criteria. It appears now that the criterion of quality has to be taken into consideration as well.

The results obtained for ferrous bars allow formulating three possible hypotheses:

- Firstly, during Antiquity, craftsmen seem to have known how to differentiate between ferritic iron, steel, or phosphoric iron. The cleansing degree was also considered.
- Secondly, the standard related to the quality of the metal was apparently recognised by the producer (smelting, depurating and compacting workshops), and also by the people who imported these products (merchants).
- Lastly, this standard also means that each type of bar – i.e. each metal quality – could be associated to functional destinations depending on the mechanical behaviour and on the length which was given by the number of blooms welded together.

Despite the significant work carried out in the present study, these first results must be confirmed by further analyses of half-products and bars, in order to ensure a better representativeness of the metallographic analyses. Thus, in the future, more work will be conducted to confirm these first observations on a larger sample set of bars from the same period, as well as on morphological types originating from other regions. This is, in our opinion, the only way to assess the quality of the metal during a given period, and to understand how the craftsmen and merchants apprehended it in the different technico-economic stages of ancient societies. Moreover, slag inclusion analyses will be continued, by examining the trace element composition, in order to allow a better discrimination of the various origins of the PPMs. Such a complex study, involving more advanced methods such as LA-ICP-MS or confocal μXRF, can become relevant only after a first approach to the major elements composition, as the one presented here.

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References

Barrouillet, V., Lamy, A., Mut, G., Pineda, J., Siret, A., 1989. Mines et fonderies antiques et médiévales du Canigou, Mines et métallurgies antiques et médiévales de la France méridionale. Recherches récentes. Journées de Perpignan (Février 1987). Université de Toulouse Le Mirail, Ville de Perpignan, Perpignan, pp. 21–29.

Blakelock, E., Martinón-Torres, M., Veldhuijzen, H., Young, T., 2009. Slag inclusions in iron objects and the quest for provenance: an experiment and a case study. Journal of Archaeological Science 36, 1745–1757.

Bonnamour, L., 2000a. Catalogue de l'exposition "Archéologie de la Saône, le fleuve gardien de la mémoire. Errance, Paris, p. 160.
Bonnamour, L., 2000b. Les ponts romains de Chalon-sur-Saône: étude préliminaire de la pile n°3. Gallia 57, 273–306.
Cambon, C., 1989. Mines et sites métallurgiques des Corbières (Aude), Mines et métallurgies antiques et médiévales de la France méridionale. Recherches récentes. Journées de Perpignan (Février 1987). Université de Toulouse Le Mirail, Ville de Perpignan, Perpignan, pp. 31–36.
Coulibaly, E., Benoit, P., Fluzin, P., 1999. Approche des techniques de traitements thermomécaniques et thermochimiques dans la mise en forme des objets chez les forgerons du Bwamu (Mali-Burkina Faso). In: Pétrequin, P., Fluzin, P., Thiriot, J., Benoit, P. (Eds.), Arts du feu et productions artisanales. Actes des XXe rencontres internationales d'Archéologie et d'histoire d'Antibes. APDCA, Antibes, pp. 143–158.
Coustures, M.-P., Béziat, D., Tollon, F., 2003. The use of trace element analysis of entrapped slag inclusions to establish ore-bar iron links: examples from two gallo-Roman iron-making sites in France (Les Martys, Montagne Noire, and Les Ferrys, Loiret). Archaeometry 45, 599–613.
Coustures, M.-P., Rico, C., Béziat, D., Djaoui, D., Long, L., Domergue, C., Tollon, F., 2006. La provenance des barres de fer romaines des Saintes-Maries-de-la-Mer (Bouches-du-Rhône). Gallia 63, 243–261.
Decombeix, P.-M., Fabre, J.-M., Tollon, F., Domergue, C., 1998. Evaluation du volume des ferriers romains du Domaine des Forges (Les Martys, Aude), de la masse de scories qu'ils renferment et de la production de fer correspondante. Revue d'Archéométrie 22, 77–90.
Decombeix, P.-M., Domergue, C., Fabre, J.-M., Gorgues, A., Rico, C., Tollon, F., Tournier, B., 2000. Réflexions sur l'organisation de la production du fer à l'époque romaine dans le bassin supérieur de la Dure, au voisinage des Martys (Aude). Gallia 57, 23–36.
Desaulty, A.M., Dillmann, P., L'héritier, M., Mariet, C., Gratuze, B., Joron, J.L., Fluzin, P., 2009. Does it come from the Pays de Bray? Examination of an origin hypothesis for the ferrous reinforcements used in French medieval churches using major and trace element analyses. Journal of Archaeological Science 36, 2445–2462.
Desaulty, A.-M., 2008. Apport des analyses chimiques multi technique à la compréhension du comportement des éléments traces dans les filières sidérurgiques anciennes. Application aux études de provenance et à la distinction des procédés. Le cas du Pays de Bray normand., Unpublished Thesis, Université de Technologie de Belfort Montbéliard, Belfort, p. 300.
Dillmann, P., L'héritier, M., 2007. Slag inclusion analyses for studying ferrous alloys employed in French medieval buildings: supply of materials and diffusion of smelting processes. Journal of Archaeological Science 34, 1810–1823.
Domergue, C., Liou, B., 1997. L'apparition de normes dans le commerce maritime romain: le cas des métaux et des denrées transportées en amphores, Mélanges C. Domergue n°1, Pallas. revue d'études antiques 46, 11–30.
Domergue, C., Serneels, V., Cauuet, B., Pailler, J.-M., Orzechowski, S., 2006. Mines et métallurgies en Gaule à la fin de l'Âge du Fer et à l'époque romaine. In: Paunier, D. (Ed.), Celtes et Gaulois, l'Archéologie face à l'histoire, 5: la Romanisation et la question de l'héritage celtique. Actes de la table ronde de Lausanne, 17–18 juin 2005, Bibracte. Centre archéologique européen, Glux-en-Glenne, pp. 131–162.
Domergue, C., 1993. Un centre sidérurgique romain de la Montagne Noire: Le Domaine des Forges (Les Martys, Aube). Revue Archéologique de la Narbonnaise, C.N.R.S., Paris 27, 477.
Dunikowski, C., Cabboi, S., 1995. La sidérurgie chez les Sénons: les ateliers celtiques et gallo-romains des Clérimois (Yonne). Maison des Sciences de l'Homme, Paris.
Fabre, J.-M., 2004. Encadré 1: la forge d'épuration des Martys. In: Mangin, M. (Ed.), Le fer. Errance, Paris, pp. 90–91.
Fluzin, P., Ploquin, A., Serneels, V., 2000. Archéométrie des déchets de production sidérurgique. Moyens et méthodes d'identification des différents éléments de la chaîne opératoire directe. Gallia 57, 101–121.
Fluzin, P., Ploquin, A., Dabosi, F., 2004. Chapitre 4: approches métallographiques et archéométriques. In: Mangin, M. (Ed.), Le fer. Errance, Paris, pp. 113–173.
Fluzin, P., 2006. Premiers résultats des études métallographiques concernant les masses de métal brutes. In: Tizzoni, M., Cuccini, C., Ruffa, M. (Eds.), Alle origini della siderurgia lecchese. Ricerche archeometallugiche ai Piani d'Erna, Lecco, pp. 129–146.
Forbes, R.-J., 1955. Studies in Ancient Technology. E.J. Brill, Leiden.
Forbes, R.-J., 1963. Studies in Ancient Technology, vol. 7. E.J. Brill, Leiden.
Forbes, R.-J., 1964. Studies in Ancient Technology, vol. 8. E.J. Brill, Leiden.
France-Lanord, A., 1963. Les lingots de fer protohistoriques. Revue d'histoire de la sidérurgie 4, 167–178.
Izard, V., Mut, G., 2007. Mines, forges et charbonnières dans le massif du Canigou. Problèmes de transition entre les industries antiques et les indices du renouveau carolingien. Domitia 8/9, 121–135.
Izard, V., 1994. Cartographie successive des entreprises métallurgiques dans les Pyrénéens nord catalanes; support préliminaire à l'étude éco-historique des forets charbonnées. Archéologie du Midi Médiéval 12, 115–129.
Laubenheimer, F., 1990. Sallèles d'Aude, un complexe de potiers gallo-romain, Documents d'Archéologie Française, vol. 26. Maison des Sciences de l'Homme, Paris, p. 157.
Laubenheimer, F., 2001. 20 ans de recherches à Sallèles d'Aude, Série Amphores. Les Belles Lettres/Presses universitaires franc-comtoises, Paris, p. 296.
Leroy, S., 2010. Circulation au Moyen Âge des matériaux ferreux issus des Pyrénées ariégeoises et de la Lombardie. Apport du couplage des analyses en éléments traces et multivariées, Unpublished Thesis, Sciences pour l'Ingénieur, Université de Technologie de Belfort-Montbéliard, Belfort, p. 390.
L'héritier, M., Dillmann, P., 2010. L'approvisionnement en fer des chantiers de construction médiévaux: coût, quantités et qualité. L'exemple des églises de la période gothique. In: Carvais, R., Guillerme, A., Nègre, V., Sakarovitch, J. (Eds.), Edifice et artifice, Histoires constructives, recueil de textes issus du Premier congrès francophone d'histoire de la construction, Paris, 19–21 juin 2008. Picard, Paris, pp. 457–466.
L'héritier, M., Dillmann, P., Benoit, P., 2010. The use of ferrous alloys for the building of gothic churches. Role, origins and production of the metal, through the examples of Rouen and Troyes. Historical Metallurgy 44, 21–35.
Long, L., Sintes, C., 2003. Commerce maritime et fluvial aux embouchures du Rhône: Le rôle d'Arles dans l'Antiquité. In: Berlanga, G.P., Ballester, J.P. (Eds.), Puertos fluviales antiguos: ciudad, desarrollo e infraestructuras (actas, IV Jornadas de arqueología subacuática, 28-30 de marzo de 2001). Universitat de València, Facultat de geografia i història, València, Universitat de València, pp. 183–201.
Long, L., Hoyau, A., Piton, J., 2002. Carte archéologique Camargue et Rhône. In: D.R.A.S.S.M (Ed.), Bilan Scientifique du Département des Recherches Archéologiques Subaquatiques et Sous-Marines 2001. Direction du Patrimoine. Sous-direction de l'Archéologie. Ministère de la Culture et de la Communication, Paris, pp. 50–55.
Long, L., Djaoui, D., Rico, C., 2005. Lingots et barres de fer: vers une typologie plus complète. In: D.R.A.S.S.M (Ed.), Bilan Scientifique du Département des Recherches Archéologiques Subaquatiques et Sous-Marines 2003. Direction du Patrimoine. Sous-direction de l'Archéologie. Ministère de la Culture et de la Communication, Paris, pp. 66–68.
Long, L., 2003. Au Large des Saintes Maries de la Mer, In: Bilan scientifique du Département des Recherches Archéologiques Subaquatiques et Sous Marines (DRASSM) 2002, pp. 51–57.
Merluzzo, P., Forrières, C., 1990. Les premières étapes du travail du fer en foyer de forge, selon des procédés traditionnels japonais. In: Benoit, P., Fluzin, P. (Eds.), Paléométallurgie du fer et cultures. Actes du Symposium international du comité pour la sidérurgie ancienne. Vulcain, Belfort, pp. 71–86.
Mut, G., Kotarba, J., 2007. Les activités métallurgiques d'époque romaine dans les Pyrénées-Orientales. In: Kotarba, J., Castellvi, G., Mazière, F. (Eds.), Carte archéologique de la Gaule: les Pyrénées-Orientales (66). Fondation Maison des Sciences de l'Homme, Paris, pp. 141–155.
Pagès, G., Long, L., Fluzin, F., Dillmann, P., 2008. Réseaux de production et standards de commercialisation du fer antique en méditerranée: les demi-produits des épaves romaines des Saintes-Maries-de-la-Mer (Bouches-du-Rhône, France). Revue Archéologique de Narbonnaise 41, 261–283. http://halshs.archives-ouvertes.fr/halshs-00420965/fr/.
Pagès, G., Fluzin, F., Long, L., 2011. L'acier, une variété de fer parmi d'autres? Approches méthodologiques des demi-produits des épaves romaines des Saintes-Maries-de-la-Mer (Bouches-du-Rhône, France). In: Dillmann, P., Hilaire-Perez, L., Verna, C. (Eds.), L'acier en Europe avant Bessemer. Actes du colloque (Paris, 2005). Méridiennes-FRAMESPA-CNRS/Université de Toulouse, Toulouse, pp. 83–110.
Pagès, G., 2010. Artisanat et économie du fer en France méditerranéenne de l'Antiquité au début du Moyen Âge: une approche interdisciplinaire. Monique Mergoil, Montagnac.
Pagès, G., in press. Un demi-produit ferreux à Vichy au IIIe s; l'apport des analyses métallographiques dans la définition de l'extension géographique et chronologique du standard des Saintes-Maries-de-la-Mer, ArcheoSciences, Revue d'Archéométrie.
Pauc, B., Pauc, P., 1998. Quelques sites de réduction du minerai de fer dans les Corbières (Aude). In: Feugère, M., Serneels, V. (Eds.), Recherches sur l'économie du fer en Méditerranée nord-occidentale. Monique Mergoil, Montagnac, pp. 116–128.
Piaskowski, J., 1984a. Proposals for a standardization of the criteria for determining technological processes in early iron and steel metallurgy. In: Scott, B. (Ed.), The crafts of the Blacksmith. Symposium pour la sidérurgie ancienne, pp. 157–168.
Piaskowski, J., 1984b. A standardization procedure for the presentation of the results of metallographic examinations of early iron implements. In: Scott, B. (Ed.), The crafts of the blacksmith. Symposium pour la sidérurgie ancienne, Belfast, pp. 169–177.
Pleiner, R., 1984. Problems in the standardisation of metallographic investigations of archaeological iron objets. In: Scott, B.G., Cleere, H. (Eds.), The Crafts of the Blacksmith, pp. 179–180. Belfast.
Rancoule, G., Solier, Y., 1977. Les mines antiques des Corbières audoises. In: Roussillon, F.H.D.L.M.E.D. (Ed.), Mines et mineurs en Languedoc et régions voisines de l'Antiquité à nos jours. Actes du XLIXe congrès de la Fédération Historique du Languedoc Méditerranéen et du Roussillon, Montpellier, pp. 23–39.
Rebiscoul, A., 2003. Le site paléométallurgique des "Ferrys" commune de La Bussière (Loiret), Revue Archéologique de Picardie, Cultivateurs, éleveurs et artisans dans les campagnes de la Gaule romaine, In: Actes du VIe colloque AGER, Compiègne (Oise), juin 2002, vol. 1/2, pp. 129–140.
Rico, C., 2005. Vingt ans de recherche sur les mines et les métallurgies romaines en péninsule ibérique (1985-2004), Pallas. revue d'études antiques 67, 217–239.
Sablayrolles, R., 1989. Six années de recherches à Saint-Denis (Aude), Mines et métallurgies antiques et médiévales de la France méridionale. Recherches récentes. Journées de Perpignan (Février 1987). Université de Toulouse Le Mirail, Ville de Perpignan, Perpignan, pp. 73–84.
Salin, E., 1952–1959. La civilisation mérovingienne d'après les sépultures, les textes et le laboratoire. A. et J. Picard et compagnie, Paris.

Schwab, R., Heger, D., Höppner, B., Pernicka, E., 2006. The provenance of iron artefacts from Manching: a multi-technique approach. Archaeometry 48, 433–452.
Scott, B.G., 1971. Applications of metallographic examination of iron artifacts to Irish Archaeology. Ulster Journal of Archaeology 34, 87–95.
Solier, Y., 1981. Les épaves de Gruissan. Archaeonautica III, 196–212.
Stewart, J.-W., Charles, J.-A., Wallach, E.-R., 2000. Iron-phosphorus-carbon system. Part I to III. Materials Science and Technology 16, 275–303.
Vega, E., Dillmann, P., Fluzin, P., 2002. Contribution à l'étude du fer phosphoreux en sidérurgie ancienne. La revue d'Archéométrie 26, 197–208.
Vega, E., Dillmann, P., L'héritier, M., Fluzin, P., Crew, P., Benoît, P., 2003. Forging of phosphoric iron. An analytical and experimental approach, Archaeometallurgy in Europe. In: Proceedings of the International Conference (Milan, 2003). Associazione Italiana di Metallurgia, Milan, pp. 337–346.
Verna, C., 2001. Le temps des Moulines. Fer, technique et société dans les Pyrénées centrales (XIIIe–XVIe siècles). Publications de la Sorbonne, Paris.